ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

20 Years and 200 Thousand Shipments . . . (see page 1)

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TECHNICAL PROGRESS REVIEWS

To meet the needs of industry for concise summaries of current atomic developments, the Atomic Energy Commission is publishing this series, Technical Progress Reviews. Issued quarterly, each of the reviews digests and evaluates the latest findings in a specific area of nuclear technology and science.

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ERRATUM

The authors of the paper "The Quality and Wholesomeness of Radiation-Pasteurized Marine Products, with Particular Reference to Fish Fillets," which begins on page 365 of *Isotopes and Radiation Technology*, Vol. 3, No. 4, should have been given as "J. W. Slavin, J. T. R. Nickerson, S. A. Goldblith, L. J. Ronsivalli, J. D. Kaylor (Supervisory Food Technologist, Bureau of Commercial Fisheries Technological Laboratory, Gloucester, Mass.), and J. J. Licciardello."

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Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development	Isotopic Power Development
Isotope Technology Development	Applications in Life Sciences
Food Irradiation	Safety
Process Radiation Development	Isotopes Around the World
Source Development	Miscellaneous

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard, D. A. Fuccillo, R. E. Greene, F. E. McKinney, and Joan E. Carden, *Editors*
A. F. Rupp, *Adviser*

Oak Ridge National Laboratory

Twentieth Anniversary of the First Commercial Isotope Shipment from Oak Ridge

On Aug. 2, 1946, Dr. Eugene P. Wigner, Research Director of Clinton Laboratories, delivered 1 mc of ^{14}C to Dr. E. V. Cowdry of Barnard Free Skin and Cancer Hospital, St. Louis, Mo., while Prescott Sandidge, Assistant Executive Director of Clinton Laboratories, and Col. Elmer E. Kirkpatrick, Deputy District Engineer of the Manhattan Engineer District, looked on (in the above photograph, left to right, are Sandidge, Wigner, Cowdry, and Kirkpatrick). With this issue of *Isotopes and Radiation Technology*, we celebrate the 20th anniversary of this event, which was the beginning of the isotope sales program at Oak Ridge National Laboratory.

Although this first shipment was a significant milestone in the development of isotope technology, it was in reality the culmination of 50 years of work by many scientists from all parts of the world. The first four articles in this issue review the history of isotopes and radiation prior to the Manhattan Project. The first of these sketches the history of radiation and isotopes from Röntgen to the discovery of fission; the second paper, by Prof. Georg Hevesy, tells of his early experiences in developing isotope techniques; Chairman Glenn T. Seaborg of the U. S. Atomic Energy Commission then describes some of the early work on the production of several radioisotopes by the cyclotron; and Dr. John H. Lawrence describes early medical uses of isotopes prepared in this way.

The next four papers describe the early days of the isotope production program in the AEC and at ORNL. In the first of these, O. M. Bizzell of the AEC tells of the development of the isotope sales program in the

Manhattan District and the AEC; A. F. Rupp and E. E. Beauchamp describe the development of methods for the commercial production of isotopes at ORNL; G. E. Boyd tells of some of the early work with isotopes in the Chemistry Division at ORNL; and S. A. Reynolds describes the development of radioisotope standards and methods for their determination.

The issue is concluded with three articles on early uses of ORNL-produced isotopes: in agriculture by C. L. Comar, in medicine by Marshall Brucer, and in industry by B. A. Fries. A statistical summary of isotope production at ORNL during the first 20 years of the program is also given.

Acknowledgment

To produce this issue has required the help and cooperation of many people. The editors especially thank the authors of the various papers for taking time out from busy schedules to prepare their papers. Dr. Arnold R. Fritsch, Special Assistant to the Chairman, provided valuable assistance with Dr. Seaborg's paper as did Elizabeth B. Anderson, Research Associate, ORINS, with Dr. Brucer's paper.

We also thank the Lawrence Radiation Laboratory; E. E. Pierce, ORNL; and J. E. Westcott, AEC, for their help with the photographs; H. F. Stringfield, ORNL, for information and files about the first shipment; J. W. Thomas, University of Pennsylvania, and Helene C. Wilson, wife of the late Dr. D. Wright Wilson, who was one of the recipients of the "second" shipment of radioisotopes, for their contribution to information about this shipment to the University of Pennsylvania School of Medicine.

The authors and editors apologize in advance to anyone who made contributions in the early days and is inadvertently omitted from these descriptions.

A Brief History of Radiation and Isotopes

The history of radiation and isotopes from the discovery of X rays by Röntgen to the discovery of fission by Meitner and Frisch is briefly reviewed in this article. This review, for convenience, is divided into three parts: in the first, the discovery of radiation and radioactivity is described; in the second, the development of methods and techniques using isotopes is discussed; and, in the third, artificial radioisotopes and nuclear fission are considered. For easy reference a chronology of these developments is included.

X Rays, Radioactivity, and Isotopes

During the last 5 years of the 19th century and the first 5 years of the 20th century, X rays and radioactivity were discovered, and by 1904 enough had been learned about these new and exciting subjects that Rutherford had written a book about them. Information in these fields was being developed so fast, however, that a year later he published, with some apologies, a revision, which constituted essentially a new book because it included so much new material and such extensive rearrangements. Rutherford summarizes the importance of this work at the beginning of the first chapter:[1]

> The close of the old and the beginning of the new century have been marked by a very rapid increase of our knowledge of that most important but comparatively little-known subject—the connection between electricity and matter. No study has been more fruitful in surprises to the investigator, both from the remarkable nature of the phenomena exhibited and from the laws controlling them. The more the subject is examined, the more complex must we suppose the constitution of matter in order to explain the remarkable effects observed. While the experimental results have led to the view that the constitution of the atom itself is very complex, at the same time they have confirmed the old theory of the discontinuous or atomic structure of matter. The study of the radio-active substances and of the discharge of electricity through gases has supplied very strong experimental evidence in support of the fundamental ideas of the existing atomic theory. It has also indicated that the atom itself is not the smallest unit of matter, but is a complicated structure made up of a number of smaller bodies.
>
> A great impetus to the study of this subject was initially given by the experiments of Lenard on the cathode rays, and by Röntgen's discovery of the X rays. An examination of the conductivity imparted to a gas by the X rays led to a clear view of the mechanism of the transport of electricity through gases by means of charged ions. This ionization theory of gases has been shown to afford a satisfactory explanation not only of the passage of electricity through flames and vapours, but also of the complicated phenomena observed when a discharge of electricity passes through a vacuum tube. At the same time, a further study of the cathode rays showed that they consisted of a stream of material particles, projected with great velocity, and possessing an apparent mass small compared with that of the hydrogen atom. The connection between the cathode and Röntgen rays and the nature of the latter were also elucidated. Much of this admirable experimental work on the nature of the electric discharge has been done by Professor J. J. Thomson and his students in the Cavendish Laboratory, Cambridge.
>
> An examination of natural substances, in order to see if they gave out dark radiations similar to X rays, led to the discovery of the radio-active bodies which possess the property of spontaneously emitting radiation, invisible to the eye, but readily detected by their action on photographic plates and their power of discharging electrified bodies. A detailed study of the radio-active bodies has revealed many new and surprising phenomena which have thrown much light, not only on the nature of the radiations themselves, but also on the processes occurring in those substances. Notwithstanding the complex nature of the phenomena, the knowledge of the subject has advanced with great rapidity, and a large amount of experimental data has now been accumulated.

RÖNTGEN AND BECQUEREL

The discovery of X rays by Röntgen in the late fall of 1895 triggered the above developments. Röntgen published the first report of his work in a relatively obscure journal;[2a] however, he sent reprints of his paper and examples of his X-ray pictures to a number of leading physicists in early January. Newspapers got the story from a physicist in Vienna and published it worldwide in early January 1896. Thus, before Röntgen first presented his results at a scientific meeting on Jan. 23, 1896, his discovery was well known to laymen as well as to scientists throughout the world. (In fact, on the same day that he was presenting his results, the journal *Nature* published an English translation of his original article and some sample X-ray pictures.[2b]) It has been estimated that over 1000 articles and 50 books were published on roentgen rays in 1896 alone.

Henri Becquerel, a French physicist, was one of those who heard about Röntgen's discovery in early 1896. Impressed by the intense fluorescence produced when cathode rays struck the glass wall in a Crookes tube, he investigated other fluorescent substances to determine if they also produced X rays. One of the fluorescent materials that Becquerel studied was potassium uranyl sulfate, which he found did give off rays similar to X rays. When he carried his studies one step further, he observed that uranium and its salts gave off these rays whether they were fluorescent or not. Becquerel announced the discovery of the radioactivity of uranium in a series of papers[3] published between February and May 1896. An interesting feature of these papers, which seem to have been published almost as soon as the work was completed, is that they lead the reader step by step through the reasoning of the author in arriving at the conclusion that radioactivity is a property of uranium and not related to the fluorescence of its salts.

MARIE AND PIERRE CURIE

Shortly after Becquerel had published his papers on the radioactivity of uranium, Marie Sklodovska Curie decided that radioactivity would be a good subject for a doctoral thesis in physics at the Sorbonne. In this work, using the electrometer that her husband Pierre and his brother Jacques had constructed and used in their discovery of piezoelectricity, she measured the ionization produced by radioactive substances. This instrument provided a convenient way to measure radioactivity in a wide variety of materials. Marie Curie's first discovery[4] (in April 1898) was that thorium was also radioactive. (This had also been discovered in Germany by Schmidt[5] in February, but his published results had not been seen in Paris.) In this paper Mme Curie also noted that certain ores of uranium, especially pitchblende, were more highly radioactive than could be accounted for by their uranium content, and she suggested that this was probably due to the presence of other radioactive elements.

At this point Pierre Curie, recognizing the importance of the work his wife was doing, dropped his own work and joined her in investigating this unknown radioactivity. Although they were working with very poor apparatus in a room that was more a storeroom than a laboratory, they brought tremendous energy and ability to the work and announced the discovery of polonium[6] in July and of radium[7] in December 1898. Following the announcement of the discovery of these elements, the Curies still had many years of work before the pure materials were isolated. One interesting feature of the paper announcing the discovery of polonium is that the words "radioactive" and "radioactivity" were used for the first time. Mme Curie describes the origin of these words in her book[8] on radioactivity published in 1910:

> C'est à cette époque qu'il a paru nécessaire de trouver un nom destiné à désigner la nouvelle propriété de la matière qui avait été découverte par H. Becquerel sur les composés d'urane, mais qui ne semblait pas appartenir exclusivement à ces composés. On peut dire que les composés d'uranium et de thorium émettent des rayons de Becquerel. J'ai nommé *radioactives* les substances qui donnent lieu à une émission de ce genre, et j'ai donné le nom de *radioactivité* à la nouvelle propriété de la matière, manifestée par ces substances. Ce nom, qui a été utilisé pour la première fois dans la publication relative à la découverte du polonium a été depuis généralement adopté.*

In this work the Curies observed some chemical effects of radiation[9] and the production of heat by radioactive decay of radium.[10]

The Curies had become so engrossed in their research that it seemed that they would never find a convenient time for Marie to stop and write a thesis; however, in early 1903 she finally wrote up the results of 5 years of research[11] and took her examination on June 25, 1903. On December 10 of the same year, she and her husband shared the Nobel prize in physics with Becquerel for the work reported in her thesis.

RUTHERFORD, SODDY, AND ASTON

While the Curies were discovering and separating new radioactive substances, Ernest Rutherford and his coworkers were studying the nature of the rays and the products that result when radioactive decay takes place. These studies, which were done over a period of many years at a number of universities, not only explained the nature of radioactive decay but also resulted in the development of the concept of a nuclear atom.

In 1899 Rutherford[12] studied the radiation from uranium and showed that there were two kinds of rays. One of these, which had little penetrating power but enormous power to ionize air, he called "alpha rays," and the other, which had more penetrating power and less ionizing power, he called "beta rays." (The next year Paul Villard[13] in France showed that radium gave off a third kind of ray that was much more penetrating than either of these and could not be deflected by a magnetic field; these were called "gamma rays.")

Shortly after the publication of the paper on the nature of the rays from uranium, Rutherford moved to McGill University (Canada), where he continued his work on the nature of radioactive decay. Rutherford was studying the decay of thorium at this time, and in many of these studies he was assisted by a young

*It was at this time that it appeared necessary to find a name to describe the new property of matter that had been discovered in uranium compounds by H. Becquerel but that did not seem to belong exclusively to these compounds. We may say that uranium and thorium compounds emit "Becquerel rays." I have called the substances which give off an emission of this kind "radioactive," and I have given the name "radioactivity" to the new property of matter shown by these substances. This name, which was used first in a publication on the discovery of polonium, has been generally adopted.

English chemist, Frederick Soddy. In 1902 Rutherford and Soddy published two papers[14] in which they showed that one element was transformed or transmuted into another by radioactive decay. (It is interesting that even this early a lack of communication between sciences seemed to exist. The original papers were published[14a] in a chemical journal because Soddy was a chemist; however, they were then rewritten and published[14b] in the *Philosophical Magazine*, which was read by physicists. In the rewriting the order of the papers was reversed.)

Rutherford continued his work with other coworkers, and by 1904 the transformation theory was relatively complete, although many details of various transformations remained to be worked out. In May 1904 Rutherford gave the Bakerian Lecture before the Royal Society and summarized the entire theory of transformation in radioactive decay.[15] In 1911 he published[16] his paper on the scattering of alpha and beta particles and the structure of the atom, from which the concept of a nuclear atom has developed.

As Rutherford, Soddy, and their coworkers studied radioactive disintegration and the transformations that occur during decay, they began to realize that some of the products could not be separated. For instance, radium D usually occurred mixed with lead when separated from pitchblende. Despite all attempts these materials could not be isolated, and they came to be known as "practically inseparable substances." As Soddy studied these substances, he came to the conclusion that they could not be separated because they were the same element. In 1913 Soddy presented his ideas at several meetings,[17] and then in late 1913 he suggested in a letter in *Nature*[18] that these elements be called "isotopes":

> So far as I personally am concerned, this has resulted in a great clarification of my ideas, and it may be helpful to others, though no doubt there is little originality in it. The same algebraic sum of the positive and negative charges in the nucleus, when the arithmetical sum is different, gives what I call "isotopes" or "isotopic elements," because they occupy the same place in the periodic table. They are chemically identical, and save only as regards the relatively few physical properties which depend upon atomic mass directly, physically identical also. Unit changes of this nuclear charge, so reckoned algebraically, give the successive places in the periodic table. For any one "place," or any one nuclear charge, more than one number of electrons in the outer-ring system may exist, and in such a case the element exhibits variable valency. But such changes of number, or of valency, concern only the ring and its external environment. There is no in- and out-going of electrons between ring and nucleus.

Although the concept of isotopes of radioactive elements was accepted at this time, the existence of isotopes of stable elements took longer to demonstrate. J. J. Thomson and F. W. Aston in 1913 obtained some evidence from positive-ray analysis that neon was a mixture of two isotopes, and they obtained an apparent density change of 0.7% when they attempted to separate them by diffusion.[19] Although these observations suggested that neon had two isotopes, the evidence was not conclusive when the work was interrupted by World War I. After the war Aston built the first mass spectrograph, with which he demonstrated the existence of a number of stable isotopes.[20]

Development of Methods Using Isotopes

The story of the development of techniques and methods for the practical use of radioisotopes is really the story of one man and his accomplishments — Georg Hevesy. Hevesy's work is too extensive to be covered in detail here; however, a collection of all his scientific papers, together with a commentary by him, was published[21] recently. At the 1955 Geneva Conference[22] and in Salzburg at the 1964 International Atomic Energy Conference on Radiochemical Methods of Analysis,[23] he gave some of his personal reminiscences about the development of these methods. Some of these highlights are covered in this issue of *Isotopes and Radiation Technology* (page 9).

The following list of significant "firsts" achieved by Hevesy and his coworkers indicates the extent of his contribution to the field of isotope technology:

- First use of an isotope as a tracer. In 1913, with Paneth, he used the lead isotope radium D as a tracer for lead in the determination of the solubilities of lead sulfide and lead chromate in water.[24] (Hevesy presented this work orally at the same meeting of the British Association in 1913 at which Soddy presented and defended his concept of isotopes.)
- First use of an isotope as a tracer in biology. In 1923 he used thorium D (an isotope of lead) to follow the movement of lead in the broad bean *(Vicia faba)*.[25]
- First use of an isotope as a tracer with an animal. In 1924, working with a dermatologist who was interested in bismuth salts for the treatment of syphilis, he studied the distribution of bismuth (radium E) in the rabbit.[26]
- First use of isotope dilution as an analytical method. In 1931, with a coworker, he used radium D to determine the lead content of a number of rocks by the isotope dilution technique.[27]
- First use of an enriched stable isotope in a biological system. In 1934, after the discovery of deuterium by Urey, Hevesy obtained some heavy water from Urey and used it to measure the mean lifetime of water in the human body[28] and the speed of exchange of water between the body of a goldfish and its environment.[29]
- First use of an artificial radioactive isotope as a tracer in biology. In 1935 Hevesy and coworkers used ^{32}P to study the metabolism of phosphorus in rats and humans. In his early work[30,31] he made ^{32}P by bombardment of sulfur with neutrons from a radium–beryllium source but later[32] used a higher specific-activity ^{32}P made by Lawrence with the University of California cyclotron.

• First use of activation analysis. In 1936 Hevesy and Levi exposed samples of rare-earth compounds to a radium–beryllium neutron source to detect and determine the impurities in them.[33]

Artificial Radioisotopes and Fission

In 1919 Rutherford showed[34] that, when an alpha particle strikes a nitrogen atom, a proton and an oxygen atom are given off. This was the first case of artificial transmutation of one element into another. In the years that followed, Rutherford and other workers demonstrated a number of nuclear transformations that occur when atoms are bombarded by various particles and rays.

Two of the most significant discoveries made in the study of nuclear transformations were the discoveries of the neutron by Chadwick[35] in 1932 and of artificial radioactivity by the Joliot-Curies[36] in 1934. Chadwick discovered the neutron by bombarding beryllium with alpha particles from polonium, and the Joliot-Curies showed that when aluminum was bombarded with alpha particles the aluminum was radioactive after the alpha source was removed.

Enrico Fermi then combined these two discoveries and showed that neutrons were ideal particles with which to bombard stable atoms to produce artificial radioactivity.[37] Using neutrons, Fermi and others made many artificial radioisotopes. The cyclotron, which was developed by Lawrence[38] beginning about 1930, also proved to be a particularly effective particle accelerator for the production of artificial radioactivity. Some of this early work is reviewed in this issue by AEC Chairman Glenn T. Seaborg (page 13) and by Dr. John H. Lawrence (page 21).

The discovery by Hahn and Strassman[39] that barium is formed when uranium is bombarded by neutrons and the explanation by Meitner and Frisch[40] of the fission of uranium led to the construction of nuclear reactors and the potential for very-large-scale production of radioisotopes. The wartime development of reactors by the United States was reviewed recently by Hewlett and Anderson[41] and earlier by Smyth[42] and will not be covered here; however, the following description by Gen. Leslie R. Groves[43] of the start of the isotope program by the Manhattan Engineer District provides a good conclusion to this review:

> During these transitional months between the war's end and the take-over by the Atomic Energy Commission, we launched a wholly new project—supplying radioactive isotopes to qualified researchers. Congressional delay in passing legislation made things particularly awkward for us in this venture, because all the work of the MED had been paid for by funds specifically appropriated for military use. Since the production of radioactive isotopes was for peaceful use, we actually had no authority to work in this area.
>
> We had always been mindful that there would be these by-products of atomic power, but we had thought their major value would be in research—medical, biological, and agricultural. We expected that they would have some industrial use, but not very much. Not long after the end of the war, however, we realized that they would probably prove to be of much greater value than we had anticipated.
>
> I felt that they should be made available for research as promptly as could be arranged. Their production would require months, since the material had first to be radiated in a pile and then reduced to the form in which it would be used. Out of my discussions with people in whom I had the utmost confidence came a final plan for radioactive isotope distribution within the United States. The first shipment was made to St. Louis, Missouri, where it was used in research on bone cancer [sic]. Soon afterward, isotopes were made available to scientists in other countries under suitable restrictions that prevented their being used for research in military areas.
>
> I did not approve of giving isotopes to researchers free of charge. In the first place, I did not think that it was in my province to give away material belonging to the United States; and secondly, I thought the material would be much more carefully used if it were paid for than if it cost nothing. In setting up the price schedules—which later proved to be excessive in some instances, too low in others—our principle was to recoup for the United States the actual operating costs of producing the isotopes, with nothing to be charged for capital investment or depreciation. While everything about this undertaking was new, including the shipping of highly radioactive material to many different locations, it did not present too many problems.
>
> Since the start of this program, the number of shipments has climbed enormously, so that in the last year the value of the radioactive isotopes to industry in this country has risen to somewhere between one-half billion and one billion dollars. It is not possible to calculate their value to research, and the surface in this area has scarcely been scratched.

The Chronology of the Development of Radiation and Radioisotopes

November 1895	W. C. Röntgen discovered X rays.[2]
February to May 1896	H. Becquerel discovered the radioactivity of uranium.[3]
1897	J. J. Thomson discovered the electron.[44]
February and April 1898	G. C. Schmidt[5] (February) and Marie Curie[4] (April) independently showed that thorium was radioactive.
July 1898	Marie and Pierre Curie discovered polonium and used the words "radioactive" and "radioactivity" for the first time.[6]
December 1898	The Curies discovered radium.[7]
January 1899	Ernest Rutherford demonstrated[12] that uranium gave off two types of radiation and called one "alpha" and the other "beta."
1899	The chemical effects of radiation were observed by the Curies.[8]
1900	Paul Villard showed that radium gave off a very penetrating ray that was not deflected by a magnetic field.[13] These rays were soon called "gamma rays."
1902	Rutherford and Frederick Soddy showed that in radioactive decay one element is transformed into another.[14]
1903	Pierre Curie and A. Laborde showed that the radioactive decay of radium produced heat.[10]
1911	Rutherford discovered the scattering of alpha and beta particles by matter and announced his theory of the nuclear atom.[16]
1913	Soddy developed the idea of the existence of isotopes among radioactive elements[17] and proposed the word "isotope" to cover this concept.[18]
1913	Hevesy and Paneth first used a radioisotope as a tracer.[24]
1919	Rutherford demonstrated the disintegration of nitrogen atoms, upon bombardment by alpha particles, to oxygen and protons.[34]
1920	F. W. Aston developed the mass spectrograph and presented positive proof of the existence of stable isotopes.[20]
1923	Hevesy first used a tracer in a biological experiment.[25]
1924	Hevesy first used a tracer in an experiment with an animal.[26]
1930	E. O. Lawrence and N. E. Edlefsen published the concept for a cyclotron.[38]
1931	Hevesy and Hobbie developed the isotopes dilution technique.[27]

1932	Chadwick discovered the neutron.[36]
1932	Urey, Brickwedde, and Murphy discovered deuterium.[45]
1934	Irène and Frédéric Joliot-Curie discovered artificial radioactivity.[36]
1934	Hevesy used an enriched stable isotope (heavy water) as a tracer in biological systems.[28,29]
1935	Hevesy used an artificial radioactive isotope, ^{32}P, in biological experiments.[30-32]
1936	Hevesy and Levi first used activation analysis.[33]
1938	Seaborg and Livingood first produced ^{131}I.
1939	Hahn, Strassman, Meitner, and Frisch discovered the fission of uranium.[39,10]
1941	Kamen and Ruben discovered ^{14}C.
Dec. 2, 1942	E. Fermi and coworkers operated the first self-sustaining chain-reacting pile.
Aug. 2, 1946	First shipment of a radioisotope from Clinton Laboratories.

(Robert H. Lafferty, Jr.)

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Radioactive Tracers and Their Applications

By Georg von Hevesy

Georg von Hevesy* was the first scientist to use natural isotopes in the study of plants and animals and also the first to use artificial ones. He introduced the use of stable isotopes and was the first to investigate creating radioactive substances in situ by neutron bombardment of samples. For 35 years he published in scientific journals, contributing more than a hundred papers.[1] Among the numerous prizes he received were the Nobel Prize in 1943 and the U. S. Atoms for Peace Award in 1959.

A Hungarian by birth, he had lived and worked in England, Denmark, the United States, Germany, and Sweden. He died at the age of 80, on July 5, 1966, just 4 weeks to the day before the 20th anniversary of the beginning of the reactor-produced isotope program. Dr. von Hevesy was asked to provide a paper for this anniversary issue of *Isotopes and Radiation Technology*, but, because of failing health, was unable to do so. The account of his early work given here is made up of extracts from two of his historical reviews.[2]

The early roots of the method of radioactive tracers are back in the Physics Department of the University of Manchester (England), which, at the time, was under the inspiring leadership of Lord, then Professor, Ernest Rutherford. He was interested in the properties of radiation emitted by various substances, including radium D, which is abundant in the lead obtained in purification of uranium ores. Many hundreds of kilograms of lead chloride prepared from pitchblende were stored in the basement of Rutherford's institute, where I met him one day in 1911. He addressed me in his usual unconventional way, telling me that if I were worth my salt I should separate radium D from all that disturbing lead. I was then a young man and therefore optimistic; I did not doubt for a moment that I would succeed. When I failed entirely, I had to conclude that radium D and lead belong to the group of "practically inseparable substances," as such substances were called prior to the coining of the word "isotope."

I tried to find consolation in the possibility of labeling lead by adding pure radium D to a known amount of lead and tracing the path of lead atoms by means of radioactivity measurements. The only institute that handled large amounts of radium emanation, and thus of radium D, at that time was the Vienna Institute of Radium Research. Late in 1912 I went to Vienna, where I was fortunate enough to meet Dr. Paneth, who had also tried extensively, and failed, to separate radium D from lead.

Radioisotopes Added as Tracers

Dr. Paneth and I worked together on tracer experiments with radioactive lead and bismuth. We first applied radioactive tracers early in 1913 to the determination of the solubility of slightly soluble salts, such as lead sulfide and lead chromate, in water, using radium D as an indicator for lead. Other applications soon followed, for example, the study of the interaction of metallic lead atoms with lead ions in a surrounding aqueous solution and the study of the interaction between the lead ions in the surface layer of solid salts and the lead ions in the solution in which the salts were immersed.

When, half a century earlier, Maxwell had been calculating the diffusion rate of gaseous oxygen into gaseous nitrogen, he assumed, to simplify the calculation, that the size and mass of nitrogen and oxygen molecules are identical. He thus calculated the diffusion rate of oxygen into oxygen, which he called "self-diffusion." By labeling lead we were able to turn this

*This biographical note is adapted from an Associated Press dispatch appearing in *The New York Times*, July 7, 1966, p. 35M.

mathematical fiction into reality and measure self-diffusion in both liquid and solid metallic lead and lead compounds. Soon after, Paneth, using labeled lead and bismuth, discovered the existence of volatile hydrides of these elements and free radicals.

Radioactive tracers have found by far their most extensive applications in the life sciences. The first such study was that of the uptake by plants of ions of lead labeled with its radioactive isotope thorium B, prepared from radiothorium isolated from thorium-containing minerals. The path of lead in the plant and the extent and speed with which lead ions are released by the plant when it is placed in a culture solution containing inactive lead salts were determined by radioactive measurements. Thus it was possible to distinguish between lead ions taken up by the plant at different times and from different sources.

With the advent of artificial radioisotopes, other elements could be labeled. The first investigation, carried out in Copenhagen shortly after the discovery of artificial radioactivity by the Joliot-Curies, was a determination of the extent to which the atoms of the mineral frame of the fully grown skeleton are replaceable. Only a few minutes after introduction of labeled phosphate into the circulation of a rat, to label the plasma phosphate, ^{32}P was found in the skeleton. If all skeletal phosphate is replaced, in time, by plasma phosphate, we ought to find 1 mg of skeletal phosphate with the same radioactivity as 1 mg of plasma phosphate. This is far from being the case. In experiments in which the radioactivity of the plasma phosphate of the rabbit was kept at a constant level by daily injections, after 50 days the activity of 1 mg of the phosphate of the soft bones was only 30% of that of the plasma phosphate, and that of the hard bones was still less. This indicates that in the lifetime of the rabbit not more than about a third of the mineral constituents of the skeleton is accessible to renewal.

The determination of the turnover rate of the numerous phosphorus compounds in the living organism has been the object of hundreds of investigations. While half the terminal phosphorus atoms of the adenosine triphosphate molecule are renewed in the course of seconds, only a few of the phosphorus atoms in deoxyribonucleic acid in the nervous system, for example, are renewed in a month; in other parts of the body the renewal rate is intermediate.

Some of the bone apatite is dissolved by plasma, and new apatite is crystallized from the liquid phase. Only part of the mineral frame is involved, and we cannot completely label a full-grown organism. We did, however, obtain completely labeled offspring from mice fed radioactive calcium during pregnancy. By shifting the newborn animals to a nonradioactive foster mother and sacrificing them at intervals for the next year or two, we found that half the maternal calcium atoms are preserved during the life of the mouse. Ninety-nine percent of the body calcium is in the skeleton, and a large fraction of this is in the inaccessible part of the bone apatite. However, if we feed the offspring on radioactive food for about 100 days, we find that in a year or two a third of the labeled calcium atoms of the skeleton are lost, the rest being incorporated in inaccessible skeletal parts. By the 11th generation, mice contain no detectable calcium atoms.

In the early days after the discovery of artificial radioactivity, radioactive tracers were prepared by the action of neutrons emitted by mixtures of radium and beryllium. When a source containing 1 g of radium was used, a few microcuries of ^{32}P of high specific activity and a few other radioactive isotopes could be prepared. When the cyclotron came into use a few years later, it became possible to obtain preparations 10^5 times as active, and radioactive isotopes of the great majority of elements became available for tracer work. These substances of high radioactivities were, with the greatest generosity, put at the disposal of workers in the field by Dr. Ernest Lawrence.

Among the great number of such studies were those of Spinks and colleagues in Canada. They added ^{32}P-labeled phosphorus fertilizers (12 kg of P_2O_5 per acre) of different types to the soil and determined both the phosphorus content of the plant and its radioactivity. If, for example, 1 mg of the plant phosphorus was half as radioactive as 1 mg of the phosphorus in the fertilizers, they concluded that half the plant phosphorus came from the fertilizer and half from the soil. They also showed that the fertilizer supplied more than 60% of the phosphorus in barley harvested in June but only about 30% of the total phosphorus in barley harvested in August. The decline in the rate of supply of fertilizer phosphorus in the later part of the season is presumably due to expansion of the root system as the plant develops so that the plant can avail itself of more of the competing soil phosphorus. Of the fertilizers investigated, ammonium phosphate was shown to be the most effective, and dicalcium phosphate, the least.

I shall mention just one more of the numerous applications of radioactive tracers in agriculture and botany: the tracing in the fertilized seed of the path of the atoms coming from the pollen. In Stockholm, pollen labeled with ^{32}P was obtained from an aspen branch that had been placed, just before the pollen formed, in a solution containing 1 mc of ^{32}P for 7 days. Female aspen blossoms were fertilized with the labeled pollen, and, in the fruits collected 3 weeks later, one seed was found to contain labeled phosphorus atoms from eight or nine pollen grains. At first this result is puzzling since seed formation requires only one fertilizing pollen, just as in the animal organism the fertilization of an egg requires only one sperm. However, in the animal the numerous sperms that do not participate in the fertilization, but are degraded in the female, reach the circulatory system and have an opportunity to participate in the fetus formation. Similarly, in plants the pollen atoms that do not participate in the fertilizing process may contribute to seed formation.

Radioisotopes can also be applied to a somewhat different field, i.e., tracing the path of ancestral atoms through different generations. The fraction of maternal

atoms replaced by atoms from the food and excreted before the offspring is born and the fraction prevented from taking part in building up of offspring are found. For example, about 55 g of sodium circulates in the extracellular fluid of an 80-kg human. Administration of a negligible weight of radiosodium momentarily labels the circulating sodium uniformly. If 4% of the administered activity is excreted the first day, we conclude that 4% of all the circulating sodium left the organism and was replaced by food sodium. Half the sodium atoms originally present are lost in about a fortnight. The atoms of the 30 g of cellular sodium also in the body are preserved longer than those of the circulating sodium, some 10 g being deeply embedded in the mineral frame of the skeleton and almost quantitatively retained during life.

With the construction of the nuclear "pile," radioactive isotopes of almost every element and of high activity became available, including tritium and ^{14}C. Radiocarbon found wide application in metabolic and related studies, as well as in dating investigations following the ingenious discovery of Libby.

I shall mention one of these studies which, though possibly not the most important, is very instructive. Chaikoff and colleagues injected into the circulatory system of the rat glucose containing a negligible weight of ^{14}C—thus labeling the circulating glucose of the blood. By determining the radioactivity of 1 mg of glucose obtained from the blood at different time intervals, they could follow the speed at which the glucose molecules present at the start of the experiment left the circulation. By collecting the exhaled CO_2 and determining its radioactivity, they could follow the rate at which the carbon of these glucose molecules was converted to CO_2. The results showed that half the glucose molecules present at the start of the experiment were no longer in the circulation after 70 min.

The exhaled air is found to be radioactive a few minutes after labeled glucose has been injected. As more and more of the glucose is burned, the activity of the air increases for the first 45 min and then decreases, along with the radioactivity of the blood. The data show that 45% of the CO_2 exhaled by the rat originates from the burning of glucose and 55% from fats and other body constituents.

Sodium, potassium, chloride, phosphate, and other ions in the blood plasma are replaced by extravascular ions at a much higher rate than is glucose. Half the sodium ions in the plasma leave the circulation in 1 or 2 min. The rapidity with which these and many other ions interchange was shown only when radioactive tracers became available. On the other hand, several days are required for proteins to be half replaced.

Very beautiful results were obtained in the study of the turnover of the circulating plasma iron, a problem of great importance for medical diagnosis. Minute amounts of iron are taken up from the food, and the study of this uptake was immensely facilitated by using radioiron as a tracer. Hahn, Whipple, and their colleagues demonstrated, shortly after cyclotron-prepared iron became available, that the uptake of iron from the intestinal tract is governed mainly by the iron requirement of the body. Tedious chemical analyses could be eliminated by feeding labeled iron and determining, after several days, the radioactivity of the red corpuscles, where most of the body iron concentrates with time. Later John Lawrence and colleagues, and numerous others, studied the rate at which labeled iron atoms in the plasma leave the circulation and are replaced by others coming from various organs and, to a minor extent, from the food.

The rate of disappearance of iron atoms from the plasma is accelerated, for example, at high altitudes, where the rate of formation of the red corpuscles is more rapid than at sea level. On the other hand, it is depressed in numerous diseases, e.g., some anemias. Lawrence and colleagues demonstrated how, with time, the iron atoms present at the start of an experiment are to a large extent in the red corpuscles of a healthy human but not in those of one suffering from myeloma.

A very effective way to interfere with hemoglobin formation, and thus with the path of the circulating plasma iron, is to expose bone to ionizing radiation. This interferes with the formation of new marrow cells and may even destroy those already present. The milieu in which hemoglobin is laid down now being absent, hemoglobin formation, and utilization of iron for this purpose, ceases.

That irradiation interferes with the formation of deoxyribonucleic acid was demonstrated first with radiophosphorus and later with radiocarbon as tracers. This interference was highly specific—protein synthesis, for example, continues in the exposed tissue. When a marrow cell—in which other constituents, but not deoxyribonucleic acid, continue to form after exposure—recovers its synthesizing capacity and divides, the division can become fatal, leading to the death of the marrow cells.

Tracers Formed in Situ

We shall now consider the case in which a tracer is produced in situ in a sample by neutron bombardment. The first activation analyses were carried out with rare-earth elements obtained from the late Auer von Welsbach, the only person who possessed pure samples of all rare-earth elements before World War II. He was interested in my hafnium studies and repeatedly invited me to spend some time with him in his castle in Carinthia. The by-products of the manufacture of the Welsbach mantle and of the cerium–iron alloys used to light cigars contained large amounts of the rare-earth elements, and on each of my visits I was presented with minute amounts of these elements. One day he told me he was willing to present me with larger amounts of one of the rare-earth elements, and, for no special reason, I chose dysprosium. After the discovery of artificial radioactivity, I exposed this dysprosium sample to the neutron stream from a mixture of 600 mg of radium, 400 mc of radon, and some

beryllium. The dysprosium became very radioactive, dysprosium being the element that shows the most intense artificial radioactivity. This observation induced me to determine the dysprosium content of an yttrium sample by exposing it to a neutron stream, which was the first activation analysis. About the same time, Prof. Luigi Rolla of the University of Florence asked me if I could determine whether 2 kg of gadolinium oxide that he had prepared contained some europium. I decided to investigate the samples by activation analysis. Fifty milligrams of each of his gadolinium fractions were wrapped in a quartz film and placed symmetrically in a paraffin block that had the neutron source in its center. The intensity of the source was not sufficient to produce much activity in gadolinium, but the europium atoms captured neutrons and formed active europium isotopes. To obtain quantitative results, we made standards from Auer's pure gadolinium oxide with known additions of his pure europium and exposed them simultaneously with Rolla's samples. G. T. Seaborg, at about the same time, showed the presence of 6 ppm of gallium in iron by activation analysis, making use of the mighty deuteron beam of the Berkeley cyclotron.

A classical example of the usefulness of activation analysis is in the determination of whether or not a fatal poisoning with arsenic was a chronic one. Arsenic has an affinity for the hair. When different parts of a hair are investigated by activation analysis, all parts should be radioactive in the case of chronic poisoning; but in the case of a single large dose, the part grown between the administration of the arsenic and death should contain most of the radioactive arsenic.

Isotopic indicators have found extended application in the field of analytical chemistry. I have mentioned only a few, and those being some whose introduction I had the opportunity of following closely.

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Early Work with Radioisotopes

By Glenn T. Seaborg

In the decade preceding 1946, Dr. Glenn T. Seaborg, who since 1961 has been Chairman of the U. S. Atomic Energy Commission, was a pioneer in demonstrating the importance and utility of cyclotron-produced isotopes for research and medical purposes. Starting in 1934 as a member of the Department of Chemistry of the University of California, Berkeley, and serving from 1946 to 1961 as director of chemical research of Lawrence Radiation Laboratory, he published numerous papers on the results of his research, many of which describe the production of new elements and isotopes—particularly in the area of the transuranium elements, where he was codiscoverer of plutonium. The editors have supplemented his comments with a section that includes portions of several of his early research publications, reprinted because of their historical interest.

The early history of radioisotopes for useful medical and biological research antedates by several years the beginning of the atomic age, which we usually associate with the 1938 discovery of nuclear fission by Otto Hahn and Fritz Strassman, with 1939 elucidations by Lise Meitner and Otto Frisch. Less than four years later, the late Enrico Fermi and his colleagues operated a reactor.

Early Cyclotron Isotopes

Prior to and during World War II, only biomedical scientists who were fortunate enough to be associated with, or have effective contacts with, universities at which there were high-energy accelerators were privileged to have radioisotopes. They were able to have only small quantities, and their supplies were somewhat uncertain because only a limited amount of cyclotron time could be made available by the physicists for producing isotopes for their medical colleagues.

I have a special personal interest in these early developments that began more than a quarter of a century ago, when I was a young researcher on the Berkeley campus of the University of California. In the mid-thirties, I was one of a group of young scientists from many fields who were attracted to the great potential of the late Ernest O. Lawrence's cyclotron. Artificial radioactivity was new in those days, and we were able to find many new radioisotopes. Personally, I was concerned most with learning more about the nucleus of the atom. But we were kept aware of the utilitarian aspects of our work by the requests our biological and medical colleagues made for new radioisotopes.

As early as 1935 these colleagues were using the radioisotopes produced in the cyclotron as tracers to make new biological explorations. On one occasion during this period, in 1938, the late Dr. Joseph G. Hamilton, one of the outstanding nuclear medical pioneers, mentioned to me the limitations on his studies of thyroid metabolism imposed by the short lifetime of the radioactive iodine tracer that was available. He was working with ^{128}I, which has a half-life of only 25 min. When he inquired about the possibility of finding another iodine isotope with a longer half-life, I asked him what value would be best for his work. He replied, "Oh, about a week." Soon after that, Jack Livingood and I synthesized and identified ^{131}I with a half-life, luckily enough, of eight days.

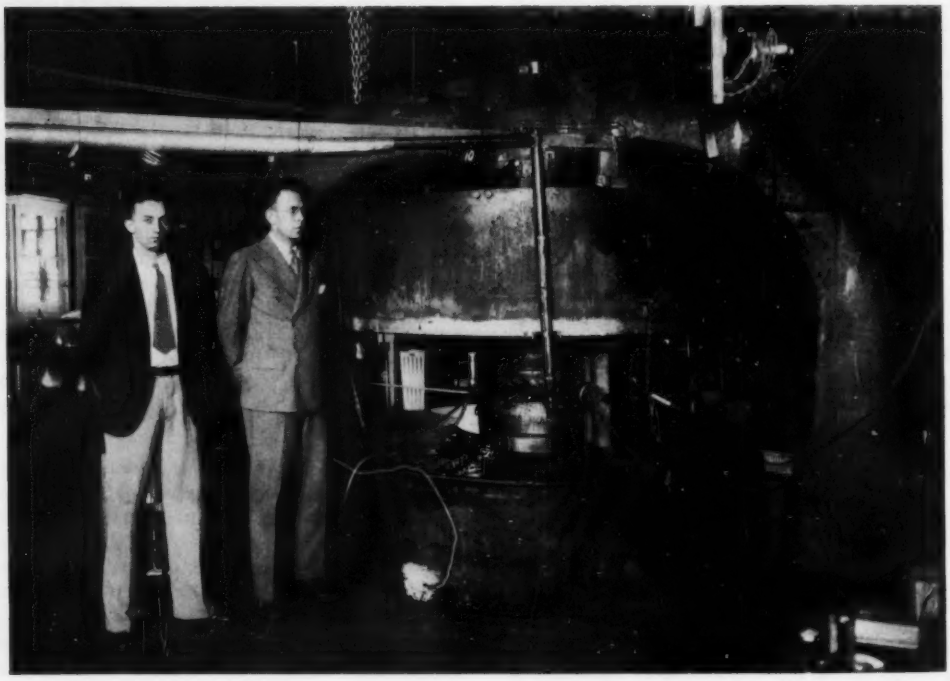
It is one of the great satisfactions of my life that ^{131}I has become one of the most useful radioisotopes for the diagnosis and treatment of disease. I am told that this isotope is now served, some half-million times a year, as the "atomic cocktail" for tracing and controlling thyroid functions. You can imagine the extent of my gratification when I tell you (for it is my pleasure to acknowledge) that my own mother has benefited from radioiodine treatment by Dr. Henry L. Jaffe at Cedars of Lebanon Hospital in Los Angeles, Calif.

Requests from other specialists in nuclear medicine led to several additional fortunate and interesting incidents of this kind in our laboratory at Berkeley.

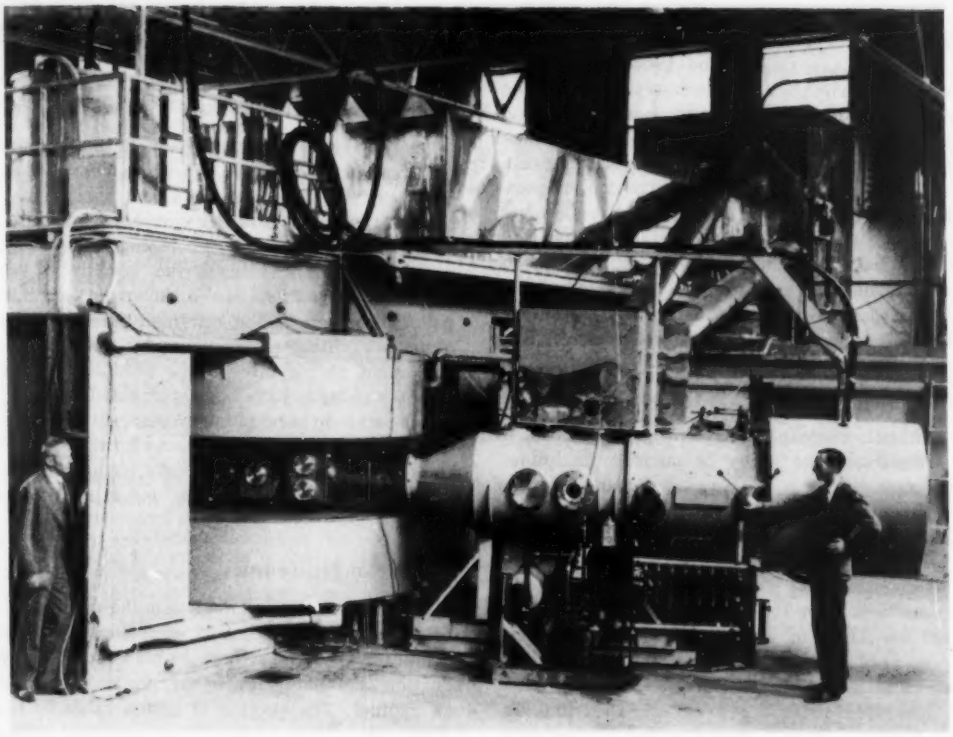
Dr. George H. Whipple, the Nobel pathologist at the University of Rochester, needed an iron isotope with a half-life of several weeks for his studies of hemoglobin formation in blood. For this use we provided ^{59}Fe, with a half-life of 45 days. I cannot claim that we in the physical sciences always fulfill the needs of our colleagues so precisely, but these examples do illustrate the importance of a close tie between the sciences.

A wartime nonmedical application of radioisotopes involved radioactive cobalt, including the 5-year ^{60}Co, whose radioactive properties we were able to characterize in the late thirties and early forties. Dr. George K. Davis, Director of Nuclear Sciences at the University of Florida, and a biologist with nuclear science leanings, was using radioactive cobalt and studying the effects of trace elements in animal and plant metabolism; but for a time he had some difficulty obtaining the cobalt he needed to make his studies. Dr. Davis appealed to Professor Robley Evans at the Massachusetts Institute of Technology who furnished him with a princely supply of radioactive cobalt, enough to provide doses for 1 cow and 300 rats, almost without charge. As a matter of fact, he was charged only $15 to take care of the time on the cyclotron. The next year, under the same arrangements, a full year's supply was made available at a cost of $30. Reactor production was "secret" until late in 1945. This is discussed in a companion article about the Manhattan Engineer District by Oscar M. Bizzell.

Most interesting is the fact that this early work at the University of Florida on trace elements in metabolism has proved to be of great significance to agriculture in the South. By these studies it was found that cattle pastured in southern Florida flourished briefly and then developed severe malnutrition because of the deficiencies of cobalt and copper and that, because of the nature of the soil, these deficiencies could not be corrected by the application of fertilizer. When copper compounds were placed in cattle feed, the situation was remedied, and a cattle industry which at one time could support only 4000 animals now supports 100,000 to 150,000 and supports them the year round instead of only for a brief fattening-up period as formerly.



Photograph taken in 1934 in the "old" Radiation Laboratory at the University of California, Berkeley. M. Stanley Livingston, now of Massachusetts Institute of Technology (left), and the late Ernest O. Lawrence stand in front of the 27-in. cyclotron. Lawrence invented the cyclotron, for which he was awarded the Nobel Prize, and founded what is now named the Lawrence Radiation Laboratory. The cyclotron in the photograph was modified in 1936 to a 37-in. machine. With this instrument, neutrons were studied, radioisotopes were created, the first artificial element, technetium, was discovered, and studies were initiated in biology and medicine with radioisotopes and heavy particles. The program of the Laboratory is supported by the Atomic Energy Commission.



The 60-in. cyclotron at the University of California Lawrence Radiation Laboratory, Berkeley, in August 1939. The machine, the most powerful atom smasher in the world at the time, had started operating early in the year. At the time of the photograph, Dr. Edwin M. McMillan was doing the work that led, a year later, to the discovery of neptunium (element 93). The instrument was used later by Dr. Glenn T. Seaborg and colleagues for the discovery of element 94 (plutonium) early in 1941. Subsequently other transuranium elements were discovered with the machine, as well as many radioisotopes, including ^{14}C. For their work Drs. Seaborg and McMillan shared the Nobel Prize in 1951. The machine was used for the "long bombardments" that produced the first weighable and visible quantities of plutonium; this material was used at Chicago by Seaborg and colleagues to work out the method for separating plutonium on an industrial scale at the Hanford, Wash., plutonium-production plant. The historic instrument was dismantled at the end of June 1962, and the magnet was sent to the Davis campus of the University of California to be converted into a more modern machine. In the photograph are Dr. Donald Cooksey, left, associate director of the Laboratory, emeritus, and Dr. G. K. Green, a member of the Laboratory staff in 1939.

Reactor Isotopes

Following World War II, as good as the prospect for useful nuclear power seemed, the most immediate application of nuclear energy lay in the exploitation of radioisotopes in many areas of potential utilization. The wartime production piles with their ready availability of large quantities of neutrons had made it possible to produce radioisotopes in greater variety and quantity than ever before. Instead of minute amounts created in cyclotron bombardment, large quantities of hundreds of radioisotopes could be produced in reactors, and powerful new sources of radiation could be developed. The list of potential uses seemed limited only by the imagination and ranged from basic research in chemistry and physics to practical applications in agriculture, industry, biology, and medicine. However, medicine accounted for most of the early uses by far.

The U.S. Atomic Energy Commission

In the United States we like to think of the Atomic Energy Act of 1946 as not only establishing the U. S. Atomic Energy Commission but also as the first major milestone in our nuclear program for the development of peaceful uses of atomic energy. Taking over in January 1947 from the procedures set up by the Manhattan District, it established a national policy that development and utilization of atomic energy should, as far as practicable, be directed toward improving the public welfare, increasing the standard of living, strengthening free competition in private enterprise, and promoting world

peace. The first Commission continued the production and distribution of isotopes—under the leadership of Dr. Paul C. Aebersold—and moved quickly to encourage the participation of industry in developing and using nuclear energy. Almost immediately, an important administrative decision was made to continue the former practice of contracting with private industrial concerns and academic institutions to perform the actual operations of the Commission's many production facilities. It is my belief, based on this experience, that the unique capacity of industry to develop new, better, and more economical ways of doing things has been one of the great assets our nation has had in nuclear-energy development.

Nuclear Medicine

Our national nuclear-energy program has also greatly stimulated the study of nuclear medicine. Beginning with the first surveys of radiation safety at the Metallurgical Laboratory in Chicago during the summer of 1942, government support expanded rapidly under a variety of Manhattan District contracts administered by Dr. Stafford L. Warren, then an Army colonel. After the newly formed Commission succeeded the Army in administering this program, the support of biomedical research continued to grow under the able direction of three men who have been closely associated with the program: Drs. Shields Warren, John C. Bugher, and Charles L. Dunham. The Commission was fortunate in that many of the physicians who had been associated with the wartime Manhattan Project, and had therefore seen at first-hand the great medical potential of nuclear energy, remained with the Commission to help develop the new tools. It is noteworthy that the first shipment of radioisotopes from the nuclear-energy project—in August 1946—went to a cancer clinic in St. Louis, Mo., for private medical research. This signaled the beginning of an era of readily available artificially produced radioisotopes at reasonable cost.

Since 1946, continuing the leads provided by the Manhattan District, the Atomic Energy Commission has carried on major efforts on four fronts to advance the cause of nuclear medicine: to produce and distribute radioactive isotopes; to provide opportunities for medical personnel to receive training in the techniques of using radioisotopes safely and effectively; to operate major medical research facilities that require specialized equipment and staff not ordinarily found in existing medical centers; and to support promising research projects through contracts with medical schools, universities, hospitals, and other research organizations.

Chiefly because of the reactor, radioisotopes are no longer novel in medical diagnosis and treatment. About 2000 hospitals and medical groups now make wide-ranging use of radioisotopes. Some 30 different radioisotopes are used in medical research, the most popular being an isotope of the most common element in biological systems, ^{14}C. Over 30 have been used for diagnostic purposes, while some half-dozen are applied in treatment.

Biology and Agriculture

The advances of recent years in biology have been heartening. Biological scientists have used great ingenuity in adapting tools and techniques of the physical sciences to their complex problems. Radioisotopes, which have given biologists eyes to "see," i.e., to locate and trace the activity of what was formerly invisible, are a foremost example. The prediction back in the thirties of the great physiologist A. V. Hill, that radioisotopes would become as important as the microscope in biological research, appears today to have been warranted.

Likewise in agriculture, the exciting early work mentioned previously has been continued and expanded to studies on fertilizer uptake, plant pathology and physiology, animal nutrition, insect studies, and soil studies.

Industrial Nucleonics

Although of later vintage than the medical, biological, and agricultural uses of isotopes, the early industrial applications likewise involved cyclotron production and naturally occurring radioisotopes such as radium. The tagging of piston rings by Ferris in 1941 with ^{32}P for lubrication and wear studies and deuteron activation of copper alloys by Sakmann, Burwell, and Irvine in 1944 for friction studies were among the early cyclotron applications. By 1947, Burwell had reported on the state of the art of isotopes in engine wear studies. Since the advent of reactor isotopes, the growth in this particular area has been remarkable.

Industrialists have perhaps been somewhat less enthusiastic about isotope usage than have medical practitioners. However, great strides have been made and large savings have been effected.

Parenthetically, I should add that private business associated with radioisotope usage has increased so much that AEC facilities now supply primarily only bulk radioisotopes for further processing and packaging by industry. Typical industry products are radiographic sources, labeled pharmaceuticals, atomic cocktails, etc.

Very large quantities of certain radioisotopes such as fission products are inherent by-products of an operating reactor and have important industrial applications. In addition, ^{238}Pu, an important step in the research leading to the discovery of fissionable ^{239}Pu, has become an extremely important isotope as a potential source of isotopic power. By the simple technique known as neutron capture, many other radioisotopes can be made. The great bulk of radioisotopes now comes from reactor operations, but the old faithful cyclotron will continue to be with us as a producer of specialty prod-

ucts. It produces radioisotopes that the reactor cannot make, it produces certain other radioisotopes more efficiently than a reactor, and it produces some isotopes with higher purity than can be achieved in a reactor. For example, during FY 1966 the ORNL 86-in. cyclotron made more than 280 irradiations for 40 customers to produce 43 different radioisotopes.

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Excerpt from *Physical Review*, 52: 135 (1937):

Radioactive Isotopes of Manganese, Iron and Cobalt

We wish to make a preliminary report on the radioactive isotopes of Mn, Fe, and Co produced when Fe is bombarded with 5.5 Mev deuterons (20-60 microampere hours) and when Co is bombarded with neutrons. The decay curves of the chemically separated fractions have been measured with a Lauritsen type electroscope.

Fe: A negative electron activity of approximately 40 days half-life is found in the Fe fraction of the Fe plus deuteron bombardment, either after precipitation (and repeated reprecipitation) as $Fe(OH)_3$ from a solution containing an excess of ammonium salts or after extraction as $FeCl_3$ by shaking with ether. An Fe activity of the same period (~40 days) has been chemically separated from a sample of CoO that was bombarded intermittently for three months with neutrons (Be + H^2) produced in the cyclotron. The conditions of this latter experiment did not allow the detection of the three day Fe isotope reported by Andersen from Co plus neutrons;[1] this isotope definitely was not produced when Fe was bombarded with deuterons.

Co: This fraction of the Fe, after deuteron bombardment, contains an 18.0 hour positron emitter, in agreement with the 18.2 hour period reported by Darling, Curtis and Cork[2] for the same reaction. In addition, there is a complex of longer periods of apparent present half-life between 100 and 200 days. Both electrons and positrons are emitted. Undoubtedly this contains the Co isotope with half-life of a year or more (reported by Sampson, Ridenour and Bleakney[3] from Co plus neutrons), and also a positron emitter of shorter period. Our work also gives support to that of Sampson and co-workers in that Co chemically separated from CoO bombarded with neutrons gives an activity which thus far is consistent with a half-life of over a year. We also find that CoO plus neutrons (without chemical separation) yields a negative electron activity with 11 minutes half-life. This period is in disagreement with the 20-minute value reported by Rotblat[4] for the same reaction but supports the values found by Heyn[5] and by Kikuchi, Takeda and Ito.[6]

Mn: The Fe plus deuteron bombardments show two new periods in the Mn precipitate; a positron emitter of about 5 days half-life and a negative electron emitter of several months period. This fraction also shows a 21-minute activity (of undetermined sign), in agreement with the 21-minute figure given by Darling, Curtis and Cork[2] for the same reaction. In disagreement with the work of Darling and co-workers the well-known 2.5-hour negative electron activity of Mn^{56} is strongly evident in this same fraction, even when very pure Fe is used for the deuteron bombardment. This is due to the reaction $Fe^{56}(n,p)Mn^{56}$, at least partially, for there is a large yield of neutrons (as observed in the cloud chamber) accompanying the transmutation of Fe to Co. This supposition is confirmed by the fact that the 2.5-hour activity also appears in a second piece of Fe placed out of the deuteron beam close to the target Fe.

A Mn positron emitter of 46 minutes half-life is produced when Cr is bombarded with deuterons.

A complete description of the work, including the decay curves and a discussion of the probable reactions involved, will be published at a later date.

We wish to express our appreciation of the continued assistance of the Chemical Foundation, the Josiah Macy, Jr. Foundation and the Research Corporation.

J. J. Livingood

Radiation Laboratory,
Department of Physics,

and

F. Fairbrother
G. T. Seaborg

Department of Chemistry, University of California,
Berkeley, California,
June 30, 1937.

[1]Andersen, Nature 138, 76 (1936).
[2]Darling, Curtis and Cork, Phys. Rev. 51, 110 (1937).
[3]Sampson, Ridenour and Bleakney, Phys. Rev. 50, 382 (1936).
[4]Rotblat, Nature 136, 515 (1935).
[5]Heyn, Physica 4, 160 (1937).
[6]Kikuchi, Takeda and Ito, Proc. Phys.-Math. Soc. Jap. 19, 43 (1937).

Excerpt from *Physical Review*, 53: 1015 (1938):

Radioactive Iodine Isotopes

We have bombarded tellurium with 8 Mev deuterons and have found two new radioactive iodine isotopes, with half-lives of 13 hours and 8 days. The latter is created by two mechanisms: (1) by the decay of radio-tellurium and (2) by direct transmutation from stable tellurium. Process (1) is demonstrated by the fact that successive extractions of iodine, from the same solution of bombarded tellurium, show a growth of the 8-day period; this must therefore be associated with either I^{129} or I^{131}, as the second activity of a double decay: $Te^{128,130}(d,p)Te^{129,131}$; $Te^{129,131} \rightarrow I^{129,131} \rightarrow Xe^{129,131}$. Process (2), which is known to occur on the basis of relative intensities, is the usual type of reaction: $Te^{128,130}(d,n)I^{129,131}$. Conclusive proof of this identification and interpretation has been furnished by the extraction of the 8-day iodine from tellurium which has been bombarded with neutrons. We cannot as yet state the period of the radio-tellurium from which this iodine grows.

Absorption measurements on the negative electrons emitted by the 8-day iodine indicate a maximum energy of 0.9 Mev; a gamma-ray is also present.

We have irradiated iodine with the fast neutrons from a lithium plus deuterons source and confirm the 13-day period reported for the same bombardment by Tape and Cork,[1] who surmised it to be due to I^{126}. We have chemically identified this activity as an iodine isotope, so that the assignment to I^{126} appears to be definite. (The antimony fraction of this same bombardment was inactive, while the tellurium precipitate exhibited a 10-hour half-life which can be ascribed definitely to Te^{127}. This tellurium period has been reported previously,[1] following neutron and deuteron bombardment of tellurium, but without definite isotopic identification.)

The yield of the 8-day iodine from $Te(d,n)I$ is very much larger than that of the strongest 13-day iodine that we have been able to produce by the reaction $I^{127}(n,2n)I^{126}$.

This research has been aided by grants from the Research Corporation, The Chemical Foundation and the Josiah Macy Jr. Foundation.

J. J. Livingood
G. T. Seaborg

Radiation Laboratory, Physics Dept. (J. J. L.)
Chemistry Department (G. T. S.),
University of California,
Berkeley, California,
June 1, 1938.

[1]G. F. Tape and J. M. Cork, Phys. Rev. 53, 676 (1938).

Excerpt from *Physical Review*, 60: 913 (1941):

Radioactive Isotopes of Cobalt

J. J. Livingood and G. T. Seaborg
Jefferson Physical Laboratory, Harvard University, Cambridge, Massachusetts, and Department of Chemistry, Radiation Laboratory, Department of Physics, University of California, Berkeley, California
December 1, 1941

The recent mass spectrographic investigation of the cobalt isotopes by Mitchell, Brown, and Fowler,[1] showing that Co^{59} is the only stable isotope of cobalt, taken together with our recently acquired transmutation data, now makes it possible for us to make isotopic assignments for all the cobalt radioactivities with almost complete certainty. Tentative assignments had been made, before all these facts were known, in our table.[2,3] The following paragraphs outline what we now believe to be the proper interpretation.

Co^{60}: *10.7 minutes and 5.3 years.* The long-lived activity has been produced by the reactions $Co^{59}(n,\gamma)Co^{60}$, $Co^{59}(d,p)Co^{60}$ and $Ni^{62}(d,\alpha)Co^{60}$. Negative beta-particles and gamma-rays are emitted and the absorption curves have already been published.[4] The short-lived activity has been produced by the reactions $Co^{59}(n,\gamma)Co^{60}$ and $Ni^{60}(n,p)Co^{60}$ and the radiation consists largely of conversion electrons. It is probable that the 10.7-minute activity constitutes an isomeric transition from an upper to a lower level in Co^{60}, and that the 5.3-year activity is due to beta-particle decay from the lower level to stable Ni^{60}.

[1]J. J. Mitchell, H. S. Brown, and R. D. Fowler, Phys. Rev. 60, 359 (1941).
[2]J. J. Livingood and G. T. Seaborg, Rev. Mod. Phys. 12, 30 (1940).
[3]G. T. Seaborg, Chem. Rev. 27, 199 (1940).
[4]J. J. Livingood and G. T. Seaborg, Phys. Rev. 53, 847 (1938).

Because security restrictions prevented publication of the wartime research in the usual way, many of the early papers were printed in a collection, the National Nuclear Energy Series. The following papers appeared in *The Transuranium Elements: Research Papers*, edited by Glenn T. Seaborg, Joseph J. Katz, and Winston M. Manning, published by McGraw-Hill Book Company, Inc., 1949. In some cases, only excerpts are included.

Paper 1.1a

A NEW ELEMENT:
RADIOACTIVE ELEMENT 94 FROM DEUTERONS ON URANIUM†

By G. T. Seaborg, E. M. McMillan, J. W. Kennedy, and A. C. Wahl

We are writing to report some results obtained in the bombardment of uranium with deuterons in the 60-in. cyclotron.

The uranium was bombarded in the form of U_3O_8, and the deuterons had to pass through a 2-mil thickness of aluminum foil before hitting the uranium target. The carefully purified element 93 fraction contained a β activity whose aluminum-absorption curve (taken on an ionization chamber connected to an FP-54 tube and also on a Lauritsen electroscope) was distinctly different from the absorption curve of a sample of the 2.3-day 93^{239} (formed from uranium plus neutrons) taken under identical conditions. The upper energy limit of the β particles from this new 93 activity is about 1 mev, compared with about 0.5 mev for 93^{239}. The ratio of γ-ray to β-particle ionization is about five times larger than for 93^{239}. The initial part of the absorption curve of this 93 from uranium plus deuterons is very similar to the initial part of the absorption curve of 93^{239}. Of course the production of 93^{239} is expected in the deuteron bombardment of uranium, from the reaction $U^{238}(d,n)93^{239}$. It is impossible to deduce from the absorption curve the relative intensities of the new 93 and of 93^{239}, since the initial parts of the individual absorption curves of these two activities might well be nearly identical. The rate of decay of the high energy β particles (0.5 to 1 mev) and γ rays from the 93 of uranium plus deuterons was determined. This gave a half life of about 2 days for the new 93. This activity is probably to be assigned to 93^{238}, 93^{236}, or 93^{235} formed in the reaction $U^{238}(d,2n)93^{238}$, $U^{235}(d,n)93^{236}$, or $U^{235}(d,2n)93^{235}$, respectively.

The growth of α particles, which might be due to the element 94 daughter of the 2-day element 93, was then looked for. We did observe the growth of α particles in the very carefully purified, as well as in the semipurified, 93 fractions, and the growth curves indicate a half life of roughly 2 days for the parent of the α emitter. The final α-particle count amounts to several hundred counts per minute for a bombardment of 200 μa-hr. This work was done with a proportional-type counter. We plan to redetermine the α-particle growth curve more accurately using an ionization chamber and linear amplifier with the help of a magnetic field to bend out the very strong β-particle background. The α particles have a range of approximately 3.9 cm in air.

This α activity is chemically separable from uranium and 93. The chemical experiments so far indicate a similarity to thorium, and the activity has not yet been separated from thorium. More chemical experiments definitely must be performed before it can be regarded as proved that the α particles are due to an isotope of element 94.

†Contribution from the Department of Chemistry and the Radiation Laboratory, University of California, Berkeley.

A report submitted Jan. 28, 1941 to Physical Review for publication as a Letter to the Editor, but voluntarily withheld from publication until later [Phys. Rev., 69: 366-367 (1946)] because of the nature of its contents, is reproduced here in its original form.

Paper 1.1b

A NEW ELEMENT:
RADIOACTIVE ELEMENT 94 FROM DEUTERONS ON URANIUM†

By G. T. Seaborg, A. C. Wahl, and J. W. Kennedy

We should like to report a few more results which we have found regarding the element 94 α radioactivity formed in the 16-mev deuteron bombardment of uranium. We sent a first report on this work in a Letter to the Editor on Jan. 28, 1941. We have in the meantime performed more experiments in order to study the chemical behavior of this α-radioactive isotope. The radioactive substance can be precipitated, in what is probably the +4 valence state, as a fluoride or iodate by using a rare earth or thorium as carrier material and as a peroxyhydrate by using thorium as carrier material. However, in the presence of the extremely strong oxidizing agent persulfate ion, $S_2O_8^{--}$, plus silver as a catalyst, this radioactive isotope is oxidized to a higher valence state which does not precipitate as a fluoride. The oxidizing agent bromate ion (BrO_3^-) is not sufficiently powerful to oxidize it to this higher valence state, and hence the radioactivity comes down as a fluoride even in the presence of bromate ion. With the help of persulfate ion it has been possible to separate quantitatively this radioactivity from thorium by using the β-active UX_1 as an indicator for thorium. These experiments make it extremely prob-

able that this α radioactivity is due to an isotope of element 94. The experiments are being continued.

†Contribution from the Department of Chemistry and the Radiation Laboratory, University of California, Berkeley.

A report submitted Mar. 7, 1941 to Physical Review for publication as a Letter to the Editor, but voluntarily withheld from publication until later [Phys. Rev., 69: 367 (1946)] because of the nature of its contents, is reproduced here in its original form.

Paper 1.2

FISSIONABLE ISOTOPE OF A NEW ELEMENT: 94^{239}†

By J. W. Kennedy, G. T. Seaborg, E. Segrè, and A. C. Wahl

We would like to report that we have observed the fission of 94^{239} with slow neutrons. The cross section for the fission of 94^{239} with slow neutrons is even larger than that of U^{235}. The cross section was determined by comparing the number of fissions obtained with a sample containing 94^{239} with the number obtained with a sample of ordinary uranium under conditions which were as identical as possible in every detail. Two separate methods were used to prove that the fissions in the 94^{239} sample could not be attributed to the presence of uranium impurity: (1) complete and identical chemical tests on the isolation of rare-earth carrier material for element 94 from nonirradiated uranium, i.e., blank tests, in which the absence of uranium impurity in the final product was established by showing the absence of uranium α particles; and (2) demonstration that the ratio of the cross section for slow neutrons to that for fast neutrons for the fission of 94^{239} is different from the same ratio for natural uranium. This latter experiment has not yet been carried out in its optimum form, and we hope to submit a later report, giving details of an improved experiment.

SUMMARY

The fission cross section of 94^{239} with slow neutrons has been determined as somewhat larger than that of U^{235}. 94^{239} was found to grow from the β decay of 93^{239} at the expected rate, corresponding to a half life of 2.3 days. The half life of 94^{239} was determined to be about 3×10^4 years. The range of the 94^{239} α particles in air (15°C) is of the order of 3.6 or 3.7 cm.

†Contribution from the Department of Chemistry and the Radiation Laboratory, University of California, Berkeley.

This report was submitted for publication on May 29, 1941, to Physical Review, but was voluntarily withheld from publication until later [Phys. Rev. 70: 555 (1946)] because of the nature of its contents.

Paper 1.4

NUCLEAR PROPERTIES OF 94^{238} AND 93^{238}†

By G. T. Seaborg, A. C. Wahl, and J. W. Kennedy

The bombardment of uranium with deuterons in the 60-in. Berkeley cyclotron produces, in addition to the 2.3-day[1] 93^{239}, a new isotope of element 93, which has a half life of 2.0 days, and β and γ radiations distinctly different from those of 93^{239}. Observation of the growth of α particles from this 2.0-day 93 led, in December 1940, to the discovery[2] of element 94, and this report describes the preparation and the measurement of the physical properties of this isotope of 94. It is this isotope, which has a half life of about 50 years and 4.1-cm α particles, which we have used for a tracer to study the chemical properties of element 94 as described in another report.[3] It is not possible at present to make a definite isotopic assignment for this activity, but the best assignment‡ from the evidence at hand seems to be 94^{235} or 94^{238}.

SUMMARY

The bombardment of uranium with deuterons in the 60-in. Berkeley cyclotron produced a new isotope of element 93 having β and γ radiations different from 93^{239}. This new β-emitting isotope of 93 decays with a half life of 2.0 days to form an isotope of 94 which decays with a half life of 50 years, emitting α particles with a range of 4.1 cm. These values place this isotope near the Geiger-Nuttall curve for the actinium family.

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1. E. M. McMillan and P. H. Abelson, Phys. Rev., 57: 1185 (1940).
2. G. T. Seaborg, E. M. McMillan, J. W. Kennedy, and A. C. Wahl, Phys. Rev., 69: 366 (1946) (submitted Jan. 28, 1941); A new element: radioactive element 94 from deuterons on uranium, Paper 1.1a, this volume; G. T. Seaborg, A. C. Wahl, and J. W. Kennedy, Phys. Rev., 69: 367 (1946) (submitted Mar. 7, 1941); A new element: radioactive element 94 from deuterons on uranium, Paper 1.1b, this volume.
3. G. T. Seaborg and A. C. Wahl, The chemical properties of elements 94 and 93, Paper 1.6, this volume (written Mar. 19, 1942); J. Am. Chem. Soc., 70: 1128 (1948).

†Contribution from the Department of Chemistry and the Radiation Laboratory, University of California, Berkeley.

This article was mailed as a secret report from Berkeley, California, to the Uranium Committee in Washington, D. C., on Mar. 21, 1942; it is reproduced here in its original form.

‡Note added at time of publication: This α radioactivity was later shown to be due to the isotope 94^{238} with a slight admixture of α radioactivity due to the isotope 94^{236}. In order to avoid confusion, this paper refers to this radioactivity as due to the isotope 94^{238}.

Paper 1.6

THE CHEMICAL PROPERTIES OF ELEMENTS 94 AND 93†

By G. T. Seaborg and A. C. Wahl

This report describes the chemical experiments that we have performed on elements 94 and 93 and the chemical properties that we have deduced from these experiments.

Although these chemical experiments have been under way for element 94 since December 1940, when element 94 was discovered,[1] and for element 93 since somewhat earlier than this, the investigation has not been a very systematic one. In the case of element 93 we have repeated most of the experiments reported by McMillan and Abelson[2] in their original publication and have confirmed their results. It has been of paramount importance to develop as quickly as possible methods of isolating in very thin precipitates elements 94 and 93 from large amounts of uranium in order to study the properties[3] of 94^{239}, and, therefore, the experiments have been of a very practical and exploratory nature. It is only recently that we have begun a systematic investigation of the chemical properties; the description of these experiments is included also.

Use has been made of the α-emitting 50-year element 94, whose production and properties are described elsewhere,[1] as the tracer for element 94. The best isotopic assignment for this seems to be 94^{235} or 94^{238}. As the tracer for element 93, the β-emitting 2.3-day 93^{239} has been used.

†Contribution from the Department of Chemistry and the Radiation Laboratory, University of California, Berkeley.

This article was prepared on Mar. 19, 1942, and was mailed as a secret report from Berkeley, Calif., to the "Uranium Committee" in Washington, D. C., on Mar. 21. The experimental work was done throughout all of 1941 and the early part of 1942.

SUMMARY

The chemical properties of elements 94 and 93 have been studied by means of the tracer technique using the radioactive isotopes 94^{238} and 93^{239}. Plutonium is suggested as the name for element 94 following the convention used in the naming of neptunium (element 93) and uranium. The chemical symbols Pu and Np are suggested for plutonium and neptunium. In the reduced states the precipitation reactions of neptunium and plutonium are similar to those of the tripositive rare earths and actinium, and of the tetrapositive cerium, thorium, and uranium. Neptunium and plutonium resemble actinium, thorium, and tetrapositive uranium in that their fluorides, iodates, and hydroxides are insoluble in water; their sulfides are soluble in acid; and their metals are electropositive. Neptunium and plutonium exhibit a higher oxidation state with properties similar to those of uranium of oxidation state +6.

Probably the reduced state of both elements has an oxidation number of +3 or +4, and the formulas of the fluorides are NpF_3 and PuF_3 or NpF_4 and PuF_4. For both elements the standard oxidation-reduction potential from the metal to the reduced ionic state is greater than +0.5 volt. For the reduced to the oxidized ionic state the standard potential for neptunium is about −1.35 volt and for plutonium, −1.0 to −1.4 volts. Probably PuO_2 and NpO_2 are the oxides formed by electrodeposition.

There is practically no resemblance in chemical properties between neptunium and plutonium and rhenium and osmium. The chemical properties of neptunium and plutonium indicate that a rare-earth-type group of elements is starting at the upper end of the periodic table, and from the present data we can just as well consider the group to be starting with actinium or thorium rather than with uranium, which has been previously suggested as the starting point.

ACKNOWLEDGMENT

S. G. English and J. W. Gofman have given us valuable help in some of the experiments described in this paper.

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Paper 1.10

SEARCH FOR ELEMENTS 94 AND 93 IN NATURE. PRESENCE OF 94^{239} IN CARNOTITE†

By C. S. Garner, N. A. Bonner, and G. T. Seaborg

1. INTRODUCTION

This report describes further attempts to find a long-lived isotope of the element 94 (as well as 93) in nature. The probable presence of 94^{239} in pitchblende concentrate, from the Great Bear Lakes region, Canada, has been reported.[1] However, the concentration of this isotope amounted to only about 1 part in 10^{14}, and it was established that there was not present as much as 1 part in 10^9 of any other isotope of element 94 or 93.

In the present work, samples of carnotite (a potassium uranium vanadate), fergusonite (a rare-earth uranium columbate-tantalate), and hatchettolite (a uranium columbate-tantalate) were subjected to chemical treatment designed[2] to separate elements 94 and 93 from members of the uranium, thorium, and actinium radioactive series, which would interfere in distinguishing the α- and β-particle radioactivity from that of the former. The final fraction from each ore, in the form of a thin sample, was tested for α and β radioactivity. These experiments indicate the probable presence of 94^{239} in carnotite. In case there might be present isotopes of element 94 or 93 which were not α- or β-particle active, tests for fissions with slow neutrons and with fast neutrons were made. Since it is likely that any isotope of element 94 or 93 would either undergo fission with slow neutrons with a cross section of the order of that of U^{235} or with fast neutrons with a cross section of the order of that of U^{238}, the presence of an amount of element 94 or 93 of the order of 1 μg could be established by such neutron-fission tests. In the present work no detectable amounts of element 94 or 93 were found by these neutron tests.

SUMMARY

Samples of carnotite, fergusonite, and hatchettolite were subjected to chemical treatment designed to isolate elements 94 and 93. No evidence for the presence of appreciable amounts of element 94 or 93 was found, and the tests with slow neutrons and with fast neutrons established the upper limits of the concentrations of these elements as 1 part in 10^{10} of carnotite, 1 part in 10^9 of fergusonite, and 1 part in 10^9 of hatchettolite. A small α-particle counting rate in the final fraction from the carnotite indicates the presence of 94^{239} in the carnotite at a concentration of about 1 part in 10^{14}. The probable presence of 94^{239} in pitchblende had previously been established, and the 94^{239} in both carnotite and pitchblende is probably formed as a result of the absorption by U^{238} of some of the neutrons emitted in the uranium spontaneous-fission process.

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1. G. T. Seaborg and M. L. Perlman, J. Am. Chem. Soc., 70: 1571 (1948); Search for elements 94 and 93 in nature; presence of 94^{239} in pitchblende, Paper 1.3, this volume.

†Contribution from the Department of Chemistry, University of California, Berkeley.

This article was issued as a secret report in connection with the Plutonium Project in August 1942. The experimental work was done at the University of California during 1942. The report is unchanged from its original form except for editing to make it conform to Plutonium Project Record usage.

Paper 19.13

NUCLEAR PROPERTIES OF U^{233}; A NEW FISSIONABLE ISOTOPE OF URANIUM†

By G. T. Seaborg, J. W. Gofman, and R. W. Stoughton

The bombardment of thorium with slow neutrons produces Th^{233} [by the reaction $Th^{232}(n,\gamma)Th^{233}$], which emits β particles and has a half life of 23.5 min. The daughter of Th^{233} is the 27.4-day β-emitting Pa^{233}, which in turn decays to U^{233}. We have measured the radioactive and fission properties of U^{233}. This isotope has a half life of about 1.2×10^5 years and emits α particles having a range of 3.1 ± 0.2 cm. Our measurements on a sample of U^{233} weighing 3.8 μg show that this isotope undergoes fission with slow neutrons with a cross section of the same order as that of U^{235}. The same result was obtained in a check experiment with another sample of U^{233} weighing 0.8 μg.

†Contribution from the Department of Chemistry and the Radiation Laboratory, University of California, Berkeley.

This paper was mailed from Berkeley, California, to the Uranium Committe in Washington, D. C., on April 14, 1942. The experimental work was done during 1941 and the early part of 1942.

Some Early Studies with the Products of Nuclear Physics

By John H. Lawrence, M.D., D.Sc.

Dr. John Lawrence, internationally renowned for his pioneering work in nuclear medicine, is Director of Donner Laboratory, Associate Director of Lawrence Radiation Laboratory, and Professor of Medical Physics at the University of California, Berkeley. Many biological and medical scientists from all parts of the world have received training and guidance in these laboratories. In 1935 he and his associates first studied the biological effects of neutrons from the newly developed cyclotron, and they pioneered radioisotopes and heavy particles in medical research, diagnosis, and therapy, leading to many advances in the understanding and treatment of several diseases. The awards and honors given Dr. Lawrence and his associates during three decades are many.

The 20th anniversary of the first large-scale shipment of reactor-produced artificial radioisotopes from Oak Ridge on Aug. 2, 1946, is an important event in the history of the many benefits atomic energy is offering to humanity. I have been asked to write briefly about some of our early work at Berkeley with cyclotron-produced artificial radioisotopes during the years 1935 to 1945. This article is not intended to be a complete survey of world literature on the subject but merely comments on some of our work here.

In 1935, when one of the relatively large cyclotrons was first producing the then unheard of quantities of radioisotopes, including those of sodium, phosphorus, and iodine, investigators in the life sciences were in the vanguard of users of these new materials. Studies by Hevesy and coworkers in physiology and biochemistry were no longer limited to those which could be carried out with naturally occurring radioisotopes[1] or with the relatively small quantities produced by radium–beryllium sources. By 1935 several of us in Berkeley were already studying the metabolic processes of animals and man in both health and disease by using radioactive tracers.[2] That summer we also initiated investigations of the biological effects of the then little understood neutrons,[3] and this work laid the groundwork for radiation-protection programs when the atomic reactors came along later. When the noted physiologist Professor A. V. Hill visited us during this early period, he remarked that radioisotopes would someday be as important to biology and medicine as the microscope. And, as predicted, the biomedical uses of radioisotopes are now so widespread that an attempt to review the whole field would be akin to an attempt to review the biomedical use of the microscope.

In 1935, when Wilson cloud chambers showed to us visually the relatively dense ionization produced by the neutrons (secondary protons) from the cyclotron,[3] we were immediately interested in investigating the biological effect of this denser ionization. One of the first experiments planned with my associate Paul Aebersold was to expose a rat to the neutron beam and then to look for changes in blood counts, body weight, etc. There was not much room for both the Victoreen chamber and the rat near the dees of the cyclotron, and a very small brass cylinder was designed to hold the animal. Everything was set, and my brother, Ernest, turned on the cyclotron beam. Three minutes later the beam was turned off since we did not have any idea of how much radiation would constitute a lethal dose, and we wanted to see what effect this short exposure would have. Everyone crowded around as we crawled in between the dees to see how the animal was faring. Imagine our surprise and alarm when we found the animal dead! Even though days later histological examination of the tissues demonstrated that the rat had died of suffocation and not of radiation, we did not advertise this knowledge. The dramatic effect was that everyone thereafter had a healthy respect for this new form of radiation which was emitted from the target during isotope production in the cyclotron. This accident may

well have been the critical event leading to caution in preventing neutron exposure here and our safety record since then. Subsequently a satisfactory animal holder was designed, and the studies were extended as planned. The important finding was that, per unit dose, neutrons had a much greater biological effect than X or gamma rays.[4,5] This finding led to the establishment of new safety standards for workers around cyclotrons and reactors, the permissible exposure to neutrons being set at one-tenth that allowed for X or gamma rays. In addition to the establishment of a radiation-protection program for the nuclear age, these studies also led to further experiments with other heavy particles and to their ultimate successful use in therapy. However, that is another story which is even now unfolding; the present story is concerned chiefly with radioisotopes in medicine.

In our early tracer studies with radiophosphorus in normal and leukemic animals, we found selective localization of ^{32}P in leukemic tissue.[6,7] This finding led to the first use of radioisotopes in therapy when, on Christmas Eve in 1937, a patient with chronic lymphatic leukemia was treated with sodium radiophosphate.[8-11] Since then therapeutic uses of injected or ingested radioisotopes have become widespread. Among them, ^{32}P, with its selective localization in bone marrow and bone, continues to play an important role in the treatment of several blood diseases. It has also been used for over 25 years in the successful control of polycythemia vera,[12] so that life expectancy of these patients is now about the same as that for treated diabetic or pernicious anemia patients.

Radioiodine was soon studied in animals and man. In the early studies of my colleague Hamilton,[13-15] small amounts of radioiodine were administered to a patient, and then a Geiger counter was placed directly over the thyroid gland to determine the thyroid uptake. This simple test indicated whether or not the person was hyperthyroid (excess uptake), hypothyroid, or normal. The diagnostic use of radioiodine in thyroid disease has since become much more sophisticated with the development of isotope scanners and cameras, but basically the principle is the same, and the use of this test is now worldwide. In addition, because of the selective localization of radioiodine in thyroid tissue, administration of radioiodine could be used to treat patients with Graves' disease or hyperthyroidism, the first patients being[16,17] treated simultaneously in Berkeley and Boston in 1941. Now, of course, thousands of such patients have been treated with reactor-produced ^{131}I throughout the world. In some cases of thyroid cancer, it is also possible to use radioiodine therapeutically by administering large doses and ablating the thyroid gland or destroying local or distant thyroid metastases. It is important to note that, after more than 25 years of experience with thousands of patients, there has been no clear-cut evidence of deleterious effects from the use of either radiophosphorus or radioiodine in therapy when used in therapeutically advisable dosages.[12,18]

Several other isotopes were used in this first decade of medical research with cyclotron-produced radioisotopes. Sodium-24 was used by Hamilton to study the circulation time in normal subjects and in patients with circulatory diseases.[19] Biological investigations with radioactive calcium and strontium were carried out by Charles Pecher, myself, and associates during this early period, demonstrating that radiostrontium, like radiocalcium, was selectively concentrated in bone and that it could be used to study skeletal metabolism and might provide a possible means for treating bone cancer.[20,21] Isotopes of iron and calcium were used to study the mineral content of milk also, and the results indicated the relation between serum iron and calcium on the content of these elements in the milk secreted thereafter.[22] Metabolic studies were carried out in 1939 with cyclotron-produced radioiron to study the metabolism in anemia, and the pioneering contributions of Whipple and associates[23] and of Austoni and Greenberg[24] were important to the new understanding of the metabolism of iron. These early studies were carried out in animals, the normal and anemic animals being sacrificed at various times after administration of radioiron and the activity of different tissues measured with Geiger counters. Soon we were using high-specific-activity cyclotron-produced ^{59}Fe, and the first qualitative kinetic study of an element in the human body using an isotope was carried out with it.[25,26] Curran,[27] a pioneer in using scintillation crystals for radiation measurements, was working in the Radiation Laboratory at that time (now called the Lawrence Radiation Laboratory), and through contact with him a well type counter was built by Anger.[28] Soon also an in vivo head containing a crystal was used on a patient,[25,26] making it possible to use much smaller doses of the radioisotope and thus to decrease the radiation dose to the patient. Today, of course, in hospitals throughout the world, sophisticated iron-kinetic studies, using reactor-produced ^{59}Fe, are carried out routinely in patients with various hematological disorders to determine the underlying cause and thus the type of treatment required in controlling the disorder.

During the early period we also carried out studies on inert-gas exchange, using many short-lived cyclotron-produced radioisotopes, including nitrogen, krypton, argon, and xenon, with particular reference to the physiology of the circulation and to the basic question of gas exchange in the body and as related to the problem of aviator's bends. The greater solubility of the so-called "noble" gases in fat than in water suggested that they might have narcotic or anesthetic properties. In a series of studies on mice, the narcotic properties of these gases were demonstrated, and in the early 1950's xenon was used successfully in human surgical anesthesia.[29-31]

The first application of a short-lived positron emitter took place in 1943, when ^{11}C was used in an aviation-medical study on the problem of carbon monoxide

poisoning.[32] Carbon-11 has a half-life of about 21 min. As soon as the labeled carbon monoxide was made in the 60-in. cyclotron at Berkeley, we dashed across to Donner Laboratory (a 2-min dash) where the experiment was performed. After we breathed the radioactive carbon monoxide that had been mixed with oxygen, the expired air was collected in special bags and analyzed immediately. A Geiger counter, placed alongside a canister containing a CO_2 absorber, was used to determine continuously the amount of labeled carbon dioxide produced. These studies showed that over a period of several hours there was no significant conversion of carbon monoxide to carbon dioxide in the human body. In fact, today labeled carbon monoxide[33] and carbon dioxide breath analyses play an important part in understanding the metabolism of important carbon compounds (hemoglobin, glucose, fats, amino acids, etc.). Labeled-CO_2 breath analysis is also used diagnostically, for example, to differentiate between folic acid and vitamin B_{12} deficiency as a cause of one type of anemia.[33–36] These early studies with short-lived radioisotopes and with positron emitters have been extended by us and many other investigators, and now there is increasing interest in their use, thus requiring the construction of cyclotrons for their production. This is going on at several centers in the world.[37]

These few examples of some of the early research in the biological and medical sciences aided by radioisotopes indicated the important roles to be played by radioisotopes in advancing knowledge in medical fields. As soon as they became readily available to investigators throughout the United States and the world through the important appearance of the atomic reactor, the area of their potential application seemed unlimited. Today, only 30 years since the initial medical work and only 20 years since the first large-scale shipment of radioisotopes from Oak Ridge National Laboratory, literally hundreds of radioisotopes are already being used (and many others are being considered for use) in research, diagnosis, and therapy. The investigations that become possible as the number of available radioisotopes increases and improved methods for their study, detection, and measurement are developed lead to our better understanding of basic biological processes in health and in disease. This knowledge, in turn, will lead to improved diagnostic procedures and to the control and successful treatment of many diseases in the future.

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Early History of Radioisotopes from Reactors

By Oscar M. Bizzell

Oscar M. Bizzell, Chief of the Technology Utilization Branch, Division of Isotopes Development, U. S. Atomic Energy Commission (AEC), has had a very long association with the AEC isotope program. He was graduated with a B. S. in chemistry from the University of North Carolina in 1942. Upon completion of military service, he was employed in the Clinton Laboratories Chemistry Division from 1946 to 1948. He began his employment with the AEC in 1948 in the Isotopes Division and has held such positions as Assistant Chief of the Radiological Safety Branch and Chief of the Technology Development Branch.

At the height of World War II, reactor radioisotopes were being produced in U. S. nuclear reactors (uranium piles), under tight security wraps, and distributed to Manhattan Engineer District (MED)—the name given to the atomic bomb project—laboratories for secret wartime projects. Despite the security, however, reactor radioisotopes were sent on a few urgent missions of mercy in the midst of the war. For example, Dr. Joseph G. Hamilton, University of California Medical School at Berkeley, had been treating leukemia cases with ^{32}P from the University cyclotron. (In fact, cyclotron-produced radioisotopes had been in use for nearly a decade.) When the cyclotron became committed to wartime transuranium research, the medical school needed to keep the treatments going. Therefore informal arrangements were made for the Oak Ridge, Tenn., atomic laboratory to produce and ship 1 curie of ^{32}P per month to Berkeley for medical purposes. Ironically, a Knoxville, Tenn., child—20 miles from the secret wartime reactor at Oak Ridge—was flown to Berkeley and treated with Oak Ridge radioisotopes.

Toward the end of the war, on the recommendation of several leading research scientists, MED began considerations for producing radioisotopes in the reactor and distributing them to nonproject laboratories in the United States. Initial discussions were almost concurrent with detonation of the first atomic bomb in New Mexico, in 1945. A series of memorandums was exchanged between the offices of Col. K. D. Nichols, MED District Engineer, Oak Ridge, Tenn., and Maj. Gen. Leslie R. Groves, Officer in Charge of the MED, Washington, D. C. These memorandums were the first *official* and *formal* steps toward the peaceful uses of atomic energy.

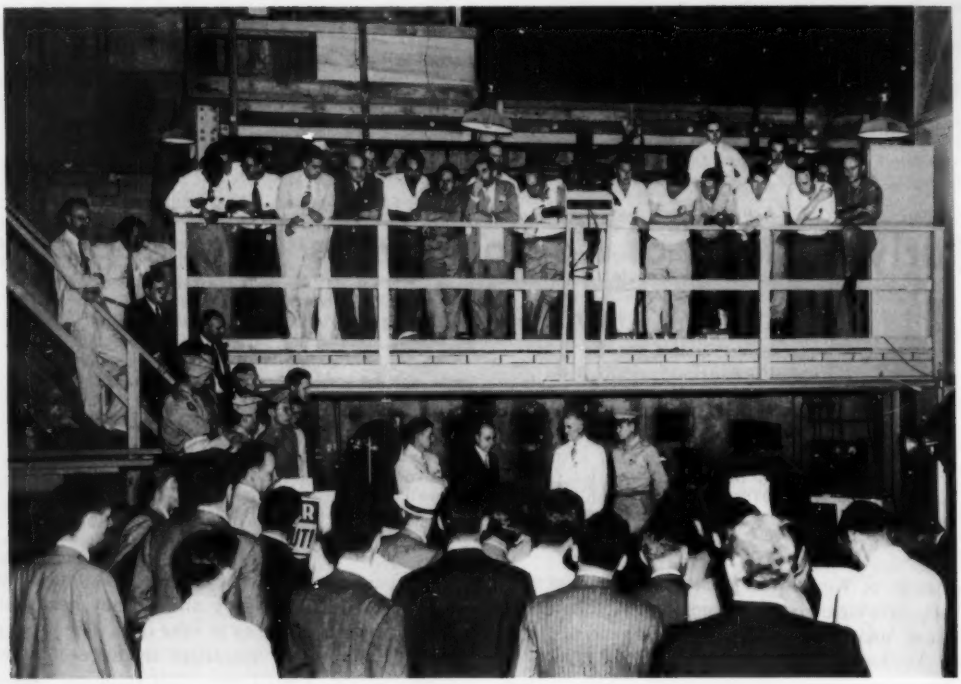
Although quantities of cyclotron isotopes available in the 1930's were very small, they heralded their tremendous usefulness in research. The prospects of virtually unlimited quantities from reactors was indeed exciting. The Clinton (Laboratories)* air-cooled pile, built for pilot-plant production of ^{239}Pu, was ideal for the production of radioactive materials.† Its production capacity was thousands of times greater than that of the most powerful cyclotron, and in a few weeks it could cheaply produce radioactive isotopes of virtually all the everyday elements. Research scientists, anticipating return to their university laboratories, were eager to gain access to this "unlimited" source of new materials. In the field of isotopes, there was no insurmountable question of policy and there were very few questions of law or economics to decide. Also, there was no question of waiting for benefits to materialize sometime in the future.

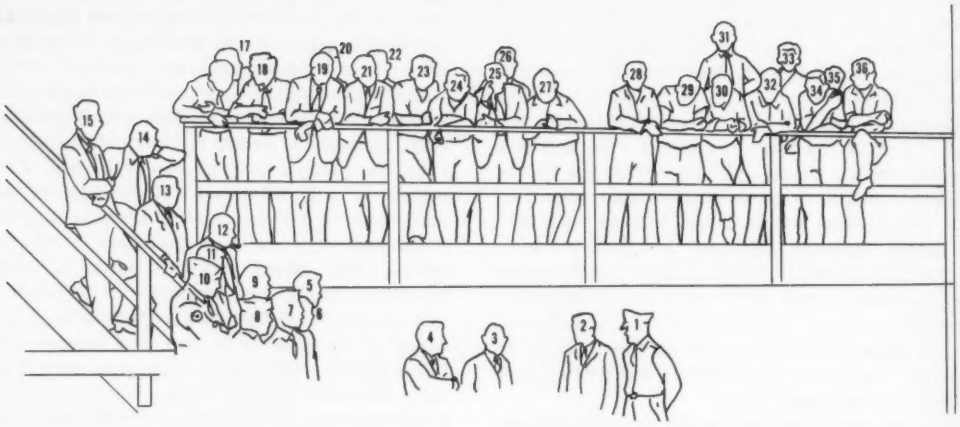
In January 1946 Dr. Paul C. Aebersold (Fig. 1) was asked to transfer from the Los Alamos Scientific Laboratory to Oak Ridge, to become chief of the Isotopes Branch in the MED Division of Research. He was a logical choice because of his experience with cyclotron radioisotopes, having been with Prof. Ernest O. Lawrence's original group. Regarding this appointment, he has written:

> When the opportunity presented itself to return to Oak Ridge in the District Research Division to help coordinate a program for distribution of radioisotopes, I was extremely happy, even though as a Californian I would have

*This facility later became Oak Ridge National Laboratory (ORNL), Oak Ridge, Tenn.

†Since this manuscript was written, the X-10 reactor has been dedicated as a national monument (November 1966).





Notable visitors, press agents, and Laboratory personnel as they witness the transfer of the first shipment of radioactive material for peacetime applications. (Twenty years ago concern was not with records, and the original photograph had no names attached. The fact that many of those pictured are no longer here and that those here cannot remember some who have left explains our inability to identify all 36 persons. Corrections and/or additions of names by readers will be appreciated by the editors.)

1. *Col. E. E. Kirkpatrick*
2. *E. V. Cowdry*
3. *Eugene P. Wigner*
4. *Prescott Sandidge*
5. *A. V. Peterson*
6. *Edward R. Tompkins*
7. *Lt. Col. W. P. Leber*
8.
9.
10.
11.
12. *Samuel Gurin*
13. *Paul Hahn*
14.
15. *Charles W. Sheppard*
16. *Ira B. Whitney*
17. *Ralph T. Overman*
18. *W. A. Rodgers*
19. *M. C. Leverett*
20. *W. Clyde Shuiten*
21. *C. R. McCullough*
22. *William H. Ray*
23. *Waldo E. Cohn*
24. *Myron B. Hawkins*
25. *Paul C. Aebersold*
26. *G. William Morgan*
27. *David O. Lintz*
28. *Alvin Barton*
29. *Delbert G. Davis*
30. *Ray W. Tucker*
31. *Henson F. Stringfield*
32. *J. D. Knox*
33. *William Redman*
34. *Charles D. Moak*
35.
36. *William J. Ladniak*

preferred the California climate. We did not mind in the least returning to Oak Ridge because our previous experience there, even though during the wartime, showed that in many ways it was a very desirable community in which to live. With the imminence of Oak Ridge as an outstanding research center of the nation, it assumed aspects that were increasingly stimulating to a scientist. Many of the plants and equipment at Oak Ridge continued to be a miracle to me. In particular, to one who was brought up scientifically on complicated electronic devices like the cyclotron, the production of radioisotopes by piling up uranium and graphite together seems "out of this world." It almost seems like giving life to inanimate material by piling it up in a certain arrangement.

It should be pointed out that I was only acting as a coordinator for the Program and that the real production effort was being done by the men in the laboratory, particularly by the Isotope groups at Clinton Laboratories. It was my job to coordinate the project supply with the nonproject demand and to try to effect understanding between all the groups involved in the detailed arrangements necessary for a production and distribution program.

Fig. 1 Dr. Paul C. Aebersold, Isotopes Branch Chief, Division of Research, MED, authorizes first peaceful distribution and use of atomic energy.

During the first 7 months of 1946, the new Isotopes Branch was a beehive of activity. Dr. Aebersold wrote memorandums for his superiors in Oak Ridge, flew to Washington to be on hand when the memorandums were delivered, and prepared replies for the return mail to Oak Ridge. This facilitated a smooth and rapid beginning for public release of these "secret" wartime materials.

Key members were added to the staff, administrative policies and procedures hammered out, application and authorization forms designed, and review committees established. The first catalog, "Availability of Radioactive Isotopes — Announcements from Headquarters, Manhattan Project, Washington, D. C.," was published in *Science*, 103: 697-705 (June 14, 1946). The introduction to the catalog stated:

> Production of tracer and therapeutic radioisotopes has been heralded as one of the great peacetime contributions of the uranium chain-reacting pile. This use of the pile will unquestionably be rich in scientific, medical, and technological applications.
>
> Manhattan Project scientific, technical, and administrative personnel have, since the inception of the pile, been cognizant of its peacetime potentialities and have, since the end of the war, been active in attempting to realize these opportunities. Since, however, war-built piles and wartime researches had other objectives, a considerable transition in researches, developments, and operations connected with piles must be effected before the supply of radioisotopes can begin to meet the demand.

The catalog also described a MED Technical Advisory Committee on Isotopes, which had been active in maintaining liaison between major laboratories of the program. This Committee included J. R. Coe, W. E. Cohn, C. R. McCullough, A. H. Snell, and K. Z. Morgan, of the Clinton Laboratories; W. H. Zinn, W. F. Libby, and R. E. Zirkle, of the Argonne and Metallurgical Laboratories; J. G. Hamilton, B. J. Moyer, and R. E. Connick, of the University of California Radiation Laboratory; J. H. Manley and R. Taschek, of the Los Alamos Scientific Laboratory; and in regard to concentrated stable isotopes, H. L. Hull and C. E. Larson, of the Tennessee Eastman Corporation, Oak Ridge.

The laboratory workers at Oak Ridge also met and solved numerous special problems before public distribution of radioisotopes could be started. Military as well as civilian personnel made contributions. For example, Corp. Myron B. Hawkins, Special Engineering Detachment, designed the first container for reactor radioisotope shipments in interstate commerce. Corporal Hawkins, after considerable deliberation, also designed the radiation symbol that is in wide use today. Thus we have the three-bladed symbol to suggest movement, but not fear, in colors initially described as magenta and heliotrope (Fig. 2).

Fig. 2 Three-bladed radiation symbol designed by Corp. Myron B. Hawkins.

Appointment of Advisory Committees

In accordance with the established custom of the Manhattan Project of seeking competent outside advice and aid on vital scientific matters, General Groves asked the president of the National Academy of Sciences to nominate a representative committee of outstanding scientists to recommend policy and aid in establishing arrangements for a desirable distribution of isotopes for diagnostic and therapeutic research that were available from Manhattan Project facilities. An interim Advisory Committee on Isotopes Distribution policy was formed, two representatives being chosen from each of the major fields of isotope application:

	Physics
Lee A. DuBridge	Chairman, Advisory Committee on Isotope Distribution
	Head, Physics Department University of Rochester
Merle A. Tuve	Head, Department of Terrestrial Magnetism Carnegie Institute of Washington
	Chemistry
Linus Pauling	Director, Gates and Crellin Chemistry Laboratories California Institute of Technology
Vincent du Vigneaud	Head, Department of Biochemistry Cornell University Medical College
	Medicine
Cornelius P. Rhoads	Director, Memorial Hospital New York City
	Chairman, Committee on Growth National Research Council
Cecil J. Watson	Head, Department of Medicine University of Minnesota Medical School
	Biology
Raymond E. Zirkle	Botany Department University of Chicago
	Director, Institute of Biophysics and Radiobiology University of Chicago
A. Baird Hastings	Head, Department of Biological Chemistry Harvard University Medical School
	Applied Science
Zay Jeffries	Manager, Chemicals Department General Electric Company Vice-President, General Electric Company
L. F. Curtiss	Chief, Radioactivity Section National Bureau of Standards

Paul C. Aebersold was appointed acting secretary to coordinate the efforts of the Committee and to effect liaison with the project.

The Committee recommendations on a suitable interim mechanism for allocation and distribution were:

1. Requests must be initiated by accredited research groups of educational institutions.

2. Requests must be initially received by the MED Isotopes Branch, which would review each request in regard to technical questions arising between the supplier and the applicant.

3. Requests would be referred to the Allocation Subcommittee, which had the responsibility of advising on the allocation and distribution of isotopes according to the scientific value of the proposed research. [The Allocation Subcommittee members were: K. T. Bainbridge (physics), Harvard University, Chairman; J. W. Kennedy (chemistry), Washington University of St. Louis; J. G. Hamilton (biology and medicine), University of California; and P. C. Aebersold (biophysics), MED, Secretary.]

4. Requests for material for use in humans would be referred to a subcommittee on Human Applications, which had final veto power on any distribution suggested for human applications. Its members were: Andrew H. Dowdy, University of Rochester, Chairman; H. L. Friedell, Western Reserve University; and G. Failla, Columbia University.

5. Small "Panels of Consultants" in a number of specialized fields of possible applications of isotopes would be appointed to offer advice as deemed necessary on scientific matters connected with requests.

6. Effective liaison would be established between the Manhattan Project technical groups and the associated advisory groups.

Licensing and Sales

In the early days close personal coordination was absolutely essential to the success of the program. Upon receipt of Form 313, Application for Radioelement, Clinton Laboratories was contacted to determine whether the type and quantity of material desired could be furnished. Concurrently, copies of the application were sent to appropriate committee members and followed up with telephone conferences. When all necessary agreements had been reached, Form 374, Radioelement Allocation Approval, was issued to the applicant. A copy of the first approval form, dated July 19, 1946, is shown in Fig. 3.

A reasonable charge was made for all isotopes to cover the "out-of-pocket" U. S. government costs resulting from the additional production necessitated by the nonproject distribution. Prices were determined on the basis of projected routine production processes. Although many isotopes were expensive to produce, especially if desired in a pure form, research-program quantities of important isotopes were not prohibitively expensive to the average scientific institution. Plans were to make materials available more rapidly and in greater quantity, as well as to reduce costs, by supplying the bombarded material when possible without subsequent chemical processing by the Manhattan Project.

In 1946 approximately 400 man-made radioactive isotopes were known, and there was at least 1 man-made radioisotope per element. The transplutonium elements curium and americium were discovered in Manhattan Project researches in 1944 and 1945, respectively. The work in connection with the Plutonium Project of the atomic bomb project had resulted in the production, or the possibility of production, of a considerable number of radioactive isotopes, and more important, this development had given rise

FORM 374	RADIOELEMENT ALLOCATION APPROVAL	SERIAL NO. 1
1. TO The Barnard Free Skin and Cancer Hospital Attn: Dr. E. V. Cowdry Dr. W. L. Simpson		2 DATE 19 July 1946
3. LOCATION 3427 Washington Avenue St. Louis 3, Missouri		

The Advisory Sub-Committee on Allocation and Distribution has approved allocation and shipment to you of material as specified below. To procure the material, it is necessary that you (1) forward a purchase order to the supplier, (2) attach to the purchase order 3 signed copies of the inclosed "Agreement and Conditions for Order and Receipt of Radioactive Materials."

The address of the supplier is: Monsanto Chemical Company, Clinton Laboratories, P. O. Box 1991, Knoxville 11, Tenn.

The supplier is authorized to bill and collect charges at fees as fixed from time to time by the Government. It is estimated that cost to you for the material and/or services will be

SPECIFICATIONS OF APPROVED RADIOISOTOPE

4. NAME OF ELEMENT	5. ATOMIC NUMBER	6. MASS	7. HALF-LIFE
Carbon	6	14	Approx. 2500 years

8. TOTAL APPROVED	9. FORM AND PURITY
1 millicurie	$BaCO_3$ approximately 3% of C radioactive

10. UNDESIRABLE CHEMICAL CONTAMINANTS
None

11. UNDESIRABLE RADIOACTIVE CONTAMINANTS
None

12. SPECIAL INSTRUCTIONS
None

APPROVED SHIPPING INSTRUCTIONS

13. AS REQUESTED BY YOU, SHIPMENT WILL BE MADE COLLECT

TO	VIA
The Barnard Free Skin and Cancer Hospital 3427 Washington Avenue St. Louis 3, Missouri	American Express Company

14. SPECIAL TIMING OF SHIPMENT OR SHIPMENTS
None

APPROVAL OF YOUR REQUEST IS BASED ON STATEMENTS CONCERNING QUANTITIES AND USES AS STATED IN YOUR "INTERIM PERIOD REQUEST FOR RADIOELEMENTS", DATED 11 July 1946

Paul C. Aebersold
Paul C. Aebersold, Chief
MANHATTAN DISTRICT, RESEARCH DIVISION, ISOTOPES BRANCH

Fig. 3 The first radioelement approval form issued to an applicant.

to vastly superior methods for the production of many isotopes. The uranium chain-reacting piles permitted transmutation of elements on a weighable scale—not only micrograms, milligrams, or grams but even kilograms in some cases.

The First Shipment—Aug. 2, 1946

All these efforts and policies were highlighted by the first shipment of radioisotopes on the afternoon of Aug. 2, 1946. Along with notable visitors and members of the press, we watched the unfolding of this historic event—certainly one of the most significant to occur at Oak Ridge. This first peacetime product of the government's huge atomic energy facilities was a pea-sized unit of ^{14}C (as $BaCO_3$), which weighed about 3 mg and contained 1 mc of radioactivity. The first shipment went to the Barnard Free Skin and Cancer Hospital, St. Louis, Mo., for research and study of the processes by which cancer is produced.

Despite its small physical size, the unit of ^{14}C for Barnard Hospital represented 100 to 1000 times as much of the isotope as had previously been made available to research in any single cyclotron-produced order. The unit was priced at $367, the actual cost to the Laboratory for production, plus handling and

shipping charges, with the total cost to the hospital of approximately $400. Dr. E. V. Cowdry and Dr. William L. Simpson, research director and associate research director of the hospital, respectively, received the millicurie of ^{14}C.

The Barnard Free Skin and Cancer Hospital workers tagged component parts of the cancer-producing molecules and then, with radiation-measuring instruments, sought to determine why different molecules produced cancer. The first step was to turn the unit of ^{14}C over to Dr. Martin K. Kamen, codiscoverer with Dr. Sam Ruben of ^{14}C in 1941. Dr. Kamen converted the ^{14}C from its carbonate form first to carbon dioxide gas and then to acetic acid. The tagged acetic acid was shipped to Dr. Paul Rothemund of the Kettering Foundation, Antioch College, Ohio, where he used it to prepare a cancer-producing agent, 20-methylcholanthrene. A part of the cancer-producing agent was retained at the Kettering Foundation for the study of its chemistry, and the remainder was used by Dr. Simpson at Barnard Hospital for the study of the artificial production of skin cancer in mice.

Several Waiting for ^{14}C

It was by chance that Barnard Hospital cleared the elaborate procurement procedure and received the first shipment. Others waiting to receive similar sized units of ^{14}C were:

Dr. D. Wright Wilson, University of Pennsylvania School of Medicine, who planned to compare sugar and lactic acid metabolism in normal and diabetic animals. He hoped to gain insight into some of the problems of diabetes by tracing ^{14}C-labeled sugar. Dr. Wilson was the recipient of the second release of Oak Ridge isotopes to the peacetime world.

Dr. James Franck, 1925 Nobel Prize winner, Professor of Physical Chemistry at the University of Chicago and world authority on photosynthesis, who wanted to use ^{14}C to study the mechanisms by which plants take energy from sunlight and store it as chemical energy.

Dr. W. D. Armstrong, Professor of Physiological Chemistry, University of Minnesota, whose investigations into the role of fluorine in the enamel of teeth were widely recognized. He planned to use ^{14}C to trace the deposition of carbon compounds in the dentin (inner pulp) and enamel of teeth and in bone.

Dr. I. L. Chaikoff, Professor of Physiology, University of California School of Medicine, who wanted to label fats with ^{14}C and study their utilization by the liver, muscle, blood, and other organs and tissues.

Other Isotopes in Demand

At the time of the first shipment, Col. Elmer E. Kirkpatrick, Deputy District Engineer, announced that hundreds of applications for radioactive isotopes, including not only ^{14}C but also many other of the 50-odd varieties producible at the Clinton Pile, had been received from the nation's leading research laboratories. He indicated that from 30 to 40 orders would be filled immediately and that several hundred additional orders were likely to be filled within the next few months.

Colonel Kirkpatrick stated that these requests for radioelements suggested widely divergent fields of scientific interest, which included the study of:

1. Mechanisms by which cancer is produced
2. Mechanisms by which plants utilize sunlight and carbon dioxide
3. Dysfunction of the thyroid gland
4. Growth and composition of teeth and bones
5. Utilization of all essential food components
6. Utilization of sugar in diabetes
7. Turnover of iron in anemic conditions
8. Absorption by plants of essential elements from soil
9. Vulcanization and polymerization of rubber
10. Problems associated with radioactive isotopes themselves

Other very early applicants for radioisotopes were:

Dr. Paul Hahn, Professor of Biochemistry, Vanderbilt University at Nashville, Tenn., requested 100 mc (or 1 unit) of radioactive gold. He planned to inject the gold in the colloidal form into the bloodstream of an animal to study the uptake of the radioactive gold by blood cells. This was a preliminary step of an investigation into possible uses of radioactive gold in certain blood diseases, such as leukemia.

Dr. John E. Christian, Professor of Pharmaceutical Chemistry, Purdue University School of Pharmacy, requested ^{32}P for use in (1) developing a new technique to test the effect of various medicinal substances on absorption of phosphate from the small intestine and (2) studying phosphorus depletion of teeth.

The American Smelting and Refining Company, Department of Agricultural Research, Salt Lake City, wanted ^{35}S to aid in a fundamental study in the metabolism of plants.

The Montefiore and Memorial Hospitals, New York City, wanted radioactive iodine, antimony, arsenic, and cesium for use in fundamental radiation studies.

It is interesting to note that in 1946 the therapeutic use of man-made isotopes was quite speculative. Some progress had been made in controlling certain forms of leukemia and polycythemia vera, both very special types of blood disorders. Only a limited number of well-qualified and experienced institutions were preparing to undertake such investigations with the newly available large quantities of reactor radioisotopes.

MED Laboratories That Assisted Oak Ridge

In 1946 it was contemplated that the radioisotopes would be prepared largely at the Clinton Laboratories, operated for MED by the Monsanto Chemical Company. However, the huge bombardment facilities of the Hanford Engineer Works at Pasco, Wash., operated by

the E. I. du Pont de Nemours & Co., Inc., for the government, and taken over by General Electric Company, Sept. 1, 1946, would also be used to the extent that the flexibility of that operation would allow. Argonne National Laboratory, announced in mid-1946 as the peacetime successor to the University of Chicago Metallurgical Laboratory, aided materially in pertinent preparations and research.

First Year of Isotope Distribution

During the first year of distribution, about 60 different reactor-produced radioisotopes were shipped to 160 institutions and organizations, representing 241 departments in 31 states and in the then Territory of Hawaii. Users in four states (California, Massachusetts, New York, and Pennsylvania) accounted for nearly half the shipments. In the first 12 months of the program, over 1100 shipments were made.

The numbers of institutions in each of the four main categories of users were approximately as follows: medical research, diagnosis, and therapy—60; educational institutions for use in research—50; industrial and profit organizations for use in both fundamental and applied research—35; and public and private nonprofit research institutions (other than educational) for use in fundamental and applied research—20.

Phosphorus-32 and ^{131}I, used chiefly in medical diagnosis and therapy, constituted the largest number of shipments of chemically separated radioisotopes. These two also accounted for the largest total activity shipped in the form of separated isotopes (over 10,000 mc each). It is particularly noteworthy that this first-year distribution included over 90 shipments of ^{14}C, with a total activity of approximately 130 mc, a quantity millions of times as great as that available for investigations before 1946.

To assist users in getting started, the Isotopes Branch established an Advisory Field Service during the first year. Isotopes Branch members visited user laboratories for first-hand review and discussion on laboratory design, safe handling practices, equipment, instrumentation, and measurement techniques. They had the authority to stop unsafe use of radioisotopes although emphasis was on education and information. To this day, 20 years later, there has been only one case of significant radiation injury from radioisotopes distributed in the United States.*

*On Mar. 29, 1960, a 33-year-old male graduate student at the University of Wisconsin, conducting research work, was accidentally exposed to a 200-curie source of ^{60}Co. Although he received a total body exposure estimated between 250–300 r and developed an acute radiation syndrome, the student is reported to have made a satisfactory recovery without specific therapy. A complete description of the accident and the results of a 15-month study that followed the exposure are given by E. C. Rossi, A. A. Thorngate, and F. C. Larson, Acute Radiation Syndrome Caused by Accidental Exposure to Cobalt 60, *J. Lab. Clin. Med.*, 50: 655-666 (1962).

At the end of the first year of distribution, a number of announcements on additional isotope distribution service had been published in the scientific journals, and popular releases on the uses of isotopes had appeared in the press and magazines. On Jan. 1, 1947, the responsibilities of the MED were transferred to the newly appointed U. S. Atomic Energy Commission, in accordance with the Atomic Energy Act of 1946. The new Commission expressed its intention to press forward the isotopes program, with a view to increasing benefits to scientific advance and human welfare.

Beginning with the *Second Semiannual Report to the Congress by the United States Atomic Energy Commission*, July 1947, a section on the Isotopes Program has been included in each issue. Detailed information on the Isotopes Program in subsequent years can be found in these reports to Congress.

Acknowledgments

The author wishes to thank the following persons for review of the manuscript and helpful comments: Dr. Willard F. Libby, University of California, Los Angeles, Dr. R. T. Overman, Oak Ridge, Tenn., Dr. G. E. Boyd, Oak Ridge National Laboratory, Dr. P. C. Aebersold, Washington, D. C., Dr. P. C. Tompkins, Federal Radiation Council, Dr. C. D. Dunham, Dr. H. D. Bruner, Dr. S. A. Lough, Mr. T. R. Jones, and Mr. J. W. Hitch, U. S. Atomic Energy Commission.

Milestones During First 10 Years of Isotopes Program

1946, Aug. 2. First public distribution of reactor-produced radioisotopes.

1947, March. Initiation of service irradiations (service whereby user submits his own sample for reactor irradiation).

1947, June. First availability of concentrated stable isotopes of ^{10}B (^{10}B complex), deuterium, and deuterium oxide (heavy water).

1947, July. First compound labeled with radiocarbon (methyl alcohol) available from AEC.

1947, September. Routine availability of chemically processed radioisotopes with standard specifications.

1947, September. Initiation of international distribution of radioisotopes. (First International Atoms for Peace Program.)

1948, January. Formation of permanent Advisory Committee on Isotope Distribution (replacing interim policy committee set up under Manhattan Project).

1948, January. Initiation of a program for distribution on a loan basis of more than 100 varieties of electromagnetically concentrated isotopes of about 30 elements.

1948, January. Enactment of regulations by the Interstate Commerce Commission for shipment of radioisotopes by common carrier (rail and truck).

1948, April. Initiation of a program making available, free of production costs, three isotopes—radioactive sodium, phosphorus, and iodine—for research, diagnosis, and therapy of cancer and allied diseases.

1948, June. Opening of first AEC-sponsored training course in basic radioisotope techniques by Oak Ridge Institute of Nuclear Studies.

1948, July. Production and distribution of radioisotope-labeled compounds begun by commercial firms.

1948, September. Announcement of availability of tritium (radioactive hydrogen) and stable ^{3}He.

1948, December. Completion of nationwide survey of ^{131}I radioactivity determinations, conducted by National Bureau of Standards and AEC, leading to program of standardization of most widely used isotopes.

1949, February. Extension of AEC's program for support of cancer research to include the making available of all normally distributed radioisotopes free of production costs for use in cancer research.

1949, June. Initiation of a program for AEC support of commercial development and synthesis of selected radioisotope-labeled compounds.

1949, July. Enactment of regulations by the Civil Aeronautics Board for shipment of radioisotopes by commercial aircraft.

1950, March. Completion of new large-scale radioisotope processing facilities at ORNL.

1950, October. Announcement of program for returning radioactive wastes to AEC facilities for disposal.

1950, December. Initiation of cooperative radiological safety program with state health departments to review procedures of radioisotope users.

1951, January. Availability of broad institutional licenses for institutions experienced in research and development with radioisotopes.

1951, March. Elimination of requirement for publication of results obtained with AEC-produced radioisotopes.

1951, April. Initiation of a program to stockpile a variety of important radioisotope-labeled compounds and make them available through ORNL.

1951, April. Publication in *Federal Register* of regulations governing radioisotopes distribution, including designation of "generally licensed" quantities (requiring no license application).

1951, July. Expansion of international distribution of radioisotopes to include all reactor-produced radioisotopes except tritium and ^{210}Po and to permit use in industrial research and application.

1951, November. First radioisotope (^{131}I) accepted by Federal Food and Drug Administration as effective new drug. Phosphorus-32 accepted June 1952 and colloidal ^{198}Au accepted October 1954.

1952, January. First film of an eight-part training-film series, "The Radioisotope," made available for promoting training in radioisotope techniques—a great aid to wider radioisotope utilization.

1952, June. Availability of licenses for use of radioisotopes by physicians in private practice.

1952, June. Revision of AEC cancer program to make more radioisotopes available at 20% of catalog prices.

1952, August. Simplification of procedures for importing radioisotopes into United States from Canada.

1953, December. Publication in *Federal Register* of criteria governing licensing of AEC radioisotopes for research and development, for human use, and for use under a broad license.

1954, July. Announcement of availability of radioisotopes in "standard reactor units."

1954, July. Revision of federal radioisotope distribution regulations to cover radioisotopes produced in any publicly and privately owned nuclear reactors located within the United States, its territories, or its possessions.

1954, July. Stable isotopes made available for international distribution.

1954, August. Enactment of Atomic Energy Act of 1954 changing certain provisions for distribution of radioisotopes.

1954, October. Appropriation approved for multikilocurie fission-product separation plant to be located at ORNL.

1955, July. Large-scale production of high-intensity ^{60}Co begun at AEC's Savannah River Plant.

1956, February. Simplification of international distribution of radioisotopes, permitting U. S. licensees to export radioisotopes of elements with atomic numbers from 3 to 83.

1956, February. Generally licensed quantities of radioisotopes increased, permitting continual use or interchange of small samples without completing an application.

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The Early Days of the Radioisotope Production Program

By A. F. Rupp and E. E. Beauchamp

A. F. Rupp, Director of the Oak Ridge National Laboratory (ORNL) Isotopes Development Center, has always been one of the strongest advocates of producing and marketing radioisotopes for peaceful uses. Mr. Rupp was graduated with a B.S. in chemical engineering from Purdue University and, after working in the chemical engineering field for several years, he joined the staff of the Manhattan Project University of Chicago Metallurgical Laboratory in 1943. Shortly thereafter he transferred to ORNL (at that time the Clinton Laboratories) and since then has been closely associated with design and engineering of many systems and processes for handling radioisotopes. His concepts for isotope processing, for waste management, and for control of contamination are in use throughout the world.

Edward E. Beauchamp has worked at Oak Ridge since 1945 when he performed fission-product analyses at Clinton Laboratories for E. I. du Pont de Nemours & Co., Inc. In 1946 he became Supervisor of the Radioisotopes Process Development Department, and in 1952 he became Superintendent of the Radioisotopes Production and Process Development Department. After several years as Superintendent of the Isotopes Sales Department at Oak Ridge National Laboratory, Mr. Beauchamp was assigned to the Isotopes Division staff in 1966 as a research engineer involved in long-range programming.

By the end of World War II, a few radioisotopes were being produced routinely at the Manhattan Project Clinton Laboratories [later Oak Ridge National Laboratory (ORNL)] as an outgrowth of the work of experimenters who needed radioisotopes for their own research. Radioisotopes made in the Graphite Reactor, some of them processed (e.g., ^{14}C, ^{131}I), were also being distributed informally to workers at other Manhattan Project sites when the decision was reached in June 1946 to make radioisotopes routinely available to off-Project research workers throughout the country. A June 1946 article in *Science* (Fig. 1) announced the availability of radioisotopes to the scientific public. An order for 1 mc of ^{14}C placed by Barnard Free Skin and Cancer Hospital, St. Louis, Mo., was officially selected as the first to be filled on Aug. 2, 1946, thus launching the highly successful radioisotope program (Fig. 2). Some of the highlights of the early production are shown in Fig. 3.

The earliest support for the fledgling radioisotope program (for needs other than nuclear research) came from scientific research and the field of medicine. The stage had been set for this work by radioisotope pioneers who used cyclotron-produced materials for tracing biochemical pathways as an aid to medical diagnosis — radioiodine for thyroid studies, radiophosphorus for blood work, and radiocarbon for metabolic investigations. The relatively enormous capacity of the Graphite Reactor for the first time enabled production of quantities of radioisotopes large enough to allow hundreds of pent-up research projects to burst forth, filling the journals within a few years with much scientific and medical information. Now, after two decades, we note that radioisotopes have been used in almost every technical field, with current emphasis on radiography, industrial process radiation, and isotopic power.

At first, work on radioisotope production at Clinton Laboratories was scattered among chemists, physicists, and engineers in many sections of the Laboratory. The first ^{14}C, for example, was made by members of the physics and chemistry groups, using an ammonium nitrate solution circulating in a loop in the pile. (In retrospect, it is surprising to note that such an advanced technique as an in-pile circulating loop, though only moderately successful at the time, should have been used so early in the radioisotope program.) Other Laboratory groups developed methods for making ^{32}P from sulfur and ^{131}I from tellurium. However, the few chemically isolated radio-

SCIENCE

Vol. 103, No. 2685 Friday, June 14, 1946

Availability of Radioactive Isotopes

Announcement From Headquarters, Manhattan Project, Washington, D.C.

PRODUCTION OF TRACER AND THERAPEUTIC RADIOISOTOPES has been heralded as one of the great peacetime contributions of the uranium chain-reacting pile. This use of the pile will unquestionably be rich in scientific, medical, and technological applications.

Manhattan Project scientific, technical, and administrative personnel have, since the inception of the pile, been cognizant of its peacetime potentialities and have, since the end of the war, been active in attempting to realize these opportunities. Since, however, war-built piles and wartime researches had other objectives, a considerable transition in researches, developments, and operations connected with piles must be effected before the supply of radioisotopes can begin to meet the demand.

AVAILABILITY OF RADIOISOTOPES

...ke the extensive variety ...ch the cyclotron becau... ...ugh greater divers... ...ling projectil...

absorbed in the introduced material that the chain reaction ceases even though the control rods are withdrawn as far as feasible. With available pile facilities, this limit does not permit the production of a sufficient quantity and quality of many radioisotopes to meet anticipated national demands. To accomplish this it would very likely be necessary to build piles especially designed for the purpose.

(5) Technical problems involved in the irradiation of some materials have been, and will continue to be, responsible for delays in making certain isotopes available by routine irradiation. Examples of such problems are: (a) proper canning of the material to prevent rupture of the container by its internal action or by the external action of the coolant, with con-

Fig. 1 Reprint of first page of article from Science, *103: (1946) announcing availability of radioactive isotopes.*

B-548 2

MONSANTO CHEMICAL COMPANY
CLINTON LABORATORIES
Oak Ridge, Tennessee

From J A. Cox SHIPPING REQUEST Radioisotope Order #CL-7-3

To Isotope Shipping Dept. Date August 1, 1946

Ship To
Dr. W. L. Simpson
Barnard Free Skin & Cancer Hospital (Research Dept
3427 Washington, St. Louis 3, Mo.
Bill To
Barnard Free Skin & Cancer Hospital (Research Dept
3427 Washington
St Louis, 3, Mo.
Spelling Billing Instructions

Customers Order # & Date
PO#842 July 22, 1946

Department No.

Route See Note

B/L No.

Container No. None

B/L Instructions

DESCRIPTION OF SHIPMENT

Quantity Ordered	Quantity Shipped	Material	Price	Extensions
1 mc C^{14}	1 mc C^{14}	_____Box, weight_____lbs.		
		Plus handling Plus Container Deposit Non-returnable container Container deposit will be refunded upon return of container, in good condition, to MCC - CL, Knoxville, Tenn. via, prepaid railway express.	None	None

Date Shipped: 8-2-46 Signed J A Cox
Isotopes Office

Shipped by: F. F. Stringfield
Isotope Shipping Department

Distribution: 1. Original to Accounting Dept.
2. Copy to Isotopes Office
3. Retained by Isotope Shipping Dept.
4. Retained by Traffic Dept
5. Producer

NOTE: Delivered to E. V. Cowdry representing Barnard Free Skin & Cancer Hospital 1 millicurie of C^{14}, August 2, 1946.

Received E. V. Cowdry
Barnard Free Skin & Can. Hosp.

Fig. 2 Document for first official shipment of radioisotopes after the Manhattan Project announcement in Science.

isotopes were not as important at the time as the providing of irradiated "units" and reactor irradiation services. Selected target materials (e.g., sodium carbonate to produce ^{24}Na) were irradiated, as well as steel piston rings, seeds, petroleum, plastics, and a myriad of other objects from false teeth to diamonds. A simple, high-purity aluminum can (with a crimped lid) containing the target material was placed in a graphite "stringer," inserted into the pile, irradiated for days or months, and sent in a shielded container to the radioisotope user. Tools to open and handle the irradiated materials also were sent to the customer.

Shortly after the first ^{14}C shipment, all radioisotope production activities were consolidated and transferred to the reactor operating group; a development "group" (consisting of the two authors of this article) was also transferred in from one of the scientific divisions. Production, shipping, and development activities were quickly organized to provide, roughly in order of importance, the following:

1. Irradiation units and services
2. Carbon-14
3. Iodine-131
4. Phosphorus-32
5. Other separated radioisotopes

Scaled-Up Production

The group was faced with the problems of devising larger scale, improved production processes for the "Big 3" (^{14}C, ^{131}I, ^{32}P) and new processes for separating and purifying some 100 other isotopes. Some order of priority was needed, and first attention was given to selecting and preparing nitrogen-containing target material for ^{14}C production [first $Ca(NO_3)_2$ and later Be_3N_2] and to developing chemical processes for separating the ^{14}C. The chemical separation process basically called for target dissolution, volatilization by transformation to $^{14}CO_2$, absorption of the $^{14}CO_2$ in alkali, and finally precipitation as $Ba^{14}CO_3$. As so often happened, one of the most difficult problems turned out to be the apparently simple operation of removing the aluminum jackets from the irradiated slugs. Another difficulty was the rigid exclusion of atmospheric CO_2 at every stage of the process. After many trials with early-day remote-control tools such as saws and modified lathes, we finally melted off the jackets in an inert-atmosphere electric-tube furnace. Typical of these early days, this work was done in a field in the open air, using distance and time limitations for radiation control. Radiation instruments were relatively primitive, but, with this and similar operations, we were able to keep radiation exposure well within the permissible limits that were in effect at that time.

Physical facilities for the radioisotope program were practically nonexistent. Any hot-laboratory space, or indeed any laboratory space, tools, and instruments that could be "scrounged" were used. Often we felt that we were following too closely in Madame Curie's footsteps! Nevertheless, this early period was challenging, exciting, and highly productive; new processes were quickly devised for ^{35}S, ^{45}Ca, ^{59}Fe, most of the important fission products, etc. By 1948 more than 50 processes had been developed, and the basic design for an isotope production area had been drawn up. Construction of the ORNL Isotope Area was completed by the end of 1949, and we were sending out 6000 shipments per year of almost 100 different kinds of radioisotope preparations.

Early Equipment

Remote-control equipment was designed and fabricated at the Laboratory and put to use for the first time on a large scale in the Isotope Area; much of it (e.g., the radioisotope packing area) served with slight change for almost 20 years and is still in use today. "Over-the-barricade-with-mirrors" was the prevailing concept, but as cells with windows and manipulators were developed during the ensuing years, they were installed in the Isotope Area's individual, highly versatile process buildings. There was a great deal of preoccupation with remote-control devices during this early period because very little equipment of this kind had been built. Each hot-laboratory man had to create his own tools. The great advances in master–slave manipulators and associated hot-cell equipment, particularly by the remote-control group at Argonne National Laboratory (ANL), were still in the future. Later, when hot-laboratory devices and instruments became available commercially, radioisotope workers were able to turn their attention to their main job with highly increased efficiency. The modern hot cell changed radioisotope processing work; next to the high-flux reactors, it is the most important improvement that has come along in radioisotope production work.

Flux Intensity

It was evident from the first that the intensity of available neutron flux was of transcendent importance in radioisotope production. While the Graphite Reactor with its 5×10^{11} $n/(cm^2)(sec)$ flux represented a mighty leap in the production capacity for radioactive materials, we had hardly started before we were wishing for 10 or 100 times that flux. Specific activities of short-lived radioisotopes were too low; hundreds of $Ca(NO_3)_2$ slugs were irradiated to produce a single curie of ^{14}C; pounds of sulfur were required to make a curie of ^{32}P; ^{60}Co sources were not nearly intense enough. We were helped in this matter quite early when a program was started with Hanford Atomic Products Operation late in 1948 to irradiate targets in their reactors, always taking into account, of course, the necessary security precautions. We were elated with the results of these irradiations and put everything we could into those

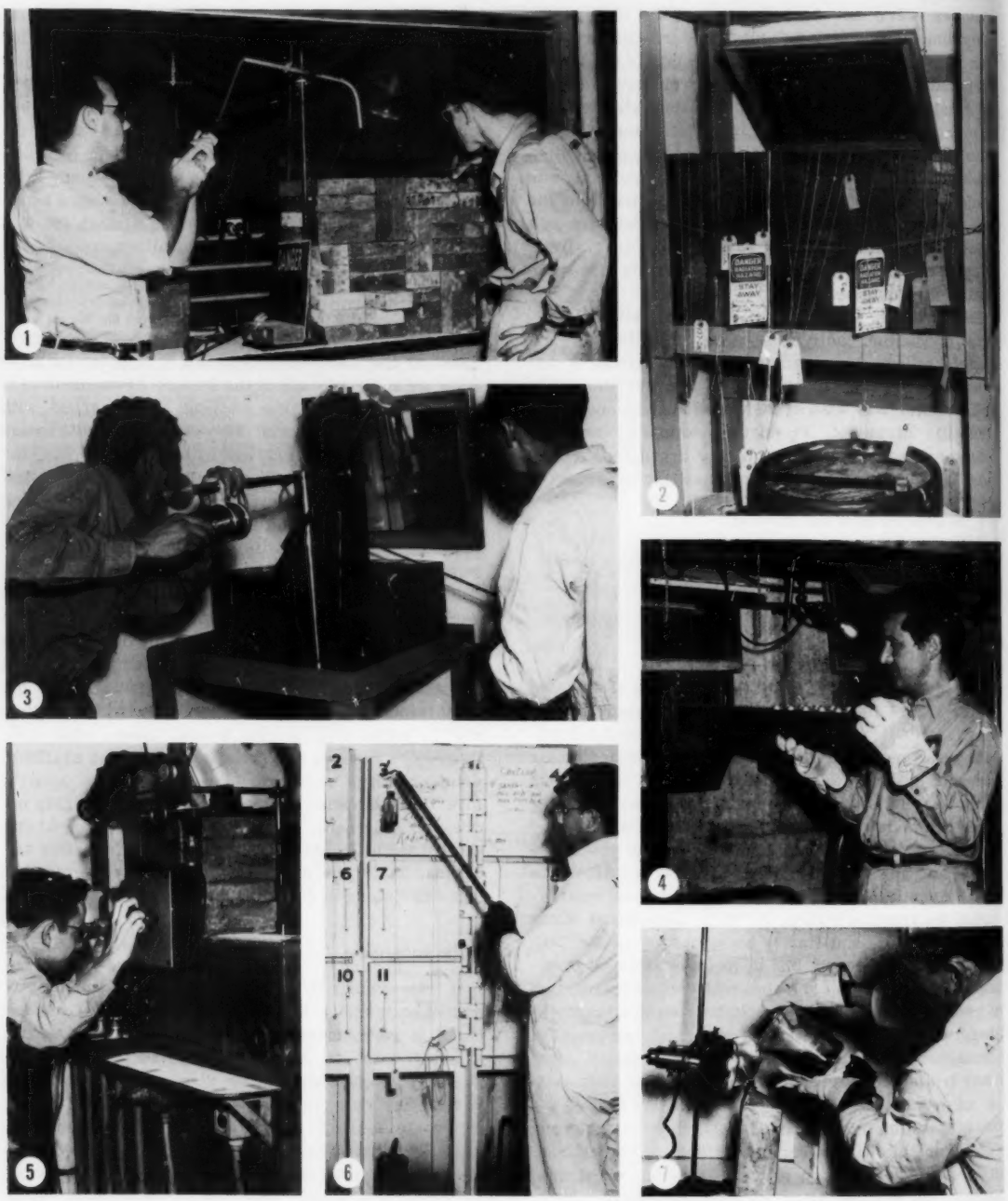

Fig. 3 Milestones and scenes of early radioisotope activities at Oak Ridge. **1.** *Radioisotope Development Laboratory.* **2.** *Storage barricade for radioisotopes awaiting shipment.* **3.** *Radioisotope dispensing equipment, 1947.* **4.** *Irradiation units in graphite stringers going into pile.* **5.** *A hot laboratory in 1946.* **6.** *Storage and handling of radioisotope analytical samples.* **7.** *Loading a sulfur can for Graphite Reactor to produce* ^{32}P. **8.** *Iodine-131 process equipment (from tellurium).* **9.** *Loading the thousandth shipment of radioisotopes, July 9, 1947, to the National Institutes of Health.* **10.** *Early cell for fission-product separations. Element 62 (Pm) was first isolated here.* **11.** *The "Argonne" manipulator cell.* **12.** *Equipment for purification of* ^{131}I *in fission-product iodine plant.* **13.** *Ground breaking for Isotope Area, 1948. Graphite Reactor building is in the background.* **14.** *Shipping containers for irradiation units and processed radioisotopes, 1948.*

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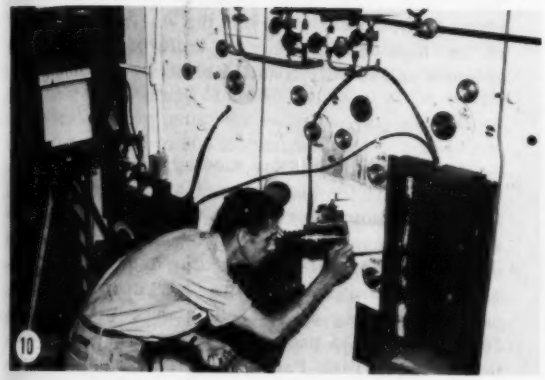
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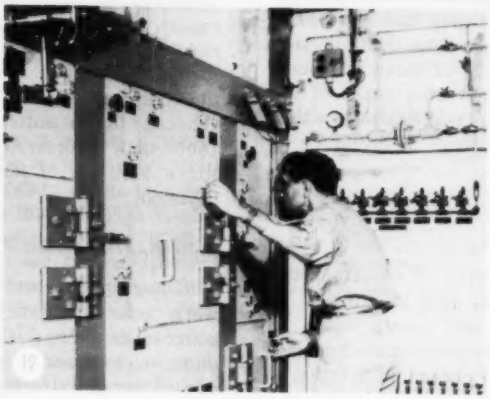
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enormous reactors as soon as possible (helped greatly by the magnificent cooperation of the Hanford technical and operating groups). Of special note was the placing in the reactor of a large charge of our newly developed ^{14}C target material, Be_3N_2, and of many shapes and sizes of cobalt metal. The Hanford reactors continue as mainstays of the radioisotopes production program to this day, being used in later years for target irradiations requiring a large flux volume. The Low Intensity Test Reactor (LITR), the progenitor of the Materials Testing Reactor (MTR), built at ORNL in 1951 primarily for hydraulic-test purposes, was converted into a 1500-kw operating unit and gave us our first high flux [$\sim 3 \times 10^{13}$ n/(cm^2)(sec)] locally. Pneumatic tubes in the LITR also helped greatly in preparing short-lived radioisotopes on fast, convenient schedules. During the first decade these three reactors enabled us to supply the bulk of the radioisotopes used throughout the world. Near the end of this period the MTR began operating, and we at last began to irradiate targets (1955) in a flux that we had only dreamed about not too many years earlier, about 2×10^{14} n/(cm^2)(sec). MTR irradiations, with those following in the Oak Ridge Research Reactor (ORR) at the Laboratory, marked the beginning of a new era in the radioisotope program.

No report would be complete without mentioning the program, even though short in years, of providing ^{14}C-labeled compounds. To make more materials available for research, the U. S. Atomic Energy Commission (AEC) distributed in the late 1940's and early 1950's ^{14}C-labeled compounds that were produced by the ORNL Chemistry Division. With the advent of commercial availability of these products, the AEC discontinued this work. This phase of the program ended with a "going-out-of-business sale" of the labeled compounds that were in inventory in 1955.

Highlights of Early Reports

On the basis of a quick review of reports of the "early days" (1946–1956), we find it difficult to single out any "most significant" points since, in those days, nearly everything accomplished appeared significant. However, we have chosen to mention a few highlights, either as anecdotes or as major accomplishments.

CYCLOTRON ISOTOPES

It is interesting that we were receiving targets from the cyclotrons at the Massachusetts Institute of Technology, University of Pittsburgh, University of California at Berkeley, and Washington University at St. Louis as early as 1948 for processing into high-quality solution-form preparations of ^{22}Na, ^{59}Fe, ^{73}As, etc. In this way radioisotopes that could not be easily produced by neutron reactions were made available to many scientists throughout the world. In 1953 we began to use the powerful ORNL 86-in. proton cyclotron for irradiating targets from which processed isotopes were prepared; later, processing service was discontinued by the AEC, and today only the unprocessed, irradiated targets from this machine are sent out.

MODERN HOT CELL

In 1952 we installed one of the first modern hot cells, complete with manipulators and viewing windows, which we dubbed the "Argonne" cell in honor of the workers at ANL who had done such a marvelous job in developing remote-control devices. The original window contained zinc bromide solution, which was subsequently replaced by a high-density glass to more nearly provide the shielding equivalent of the high-density concrete walls of the cell. The Argonne cell is still in daily service.

FISSION-PRODUCT ^{137}Cs

One of the interesting sidelights is the development of the ^{137}Cs (fission product) program. Late in 1948 the National Bureau of Standards (NBS) wanted a 10-curie source of ^{137}Cs. We had been interested in ^{137}Cs earlier but had only time to do some literature and laboratory work. The NBS source was prepared by setting up a cesium alum process in a 50-gal tank on a pad, shielded by concrete blocks, in the ORNL Tank Farm Area. The original equipment was so crude that the agitator in the tank was hardly more than a remote-control hand-operated paddle. The completed 10-curie source was delivered to the NBS in 1950. From this beginning we improved the equipment's scale and versatility, and in 1954 we reached the significant milestone of producing a 1500-curie teletherapy source for use at the Oak Ridge Institute of Nuclear Studies (ORINS) Medical Division.

LARGE AND INTENSE SOURCES

Radiographers and medical radiation therapists were interested very early in obtaining intense sources of ^{60}Co, ^{137}Cs, and ^{192}Ir to supplement radium sources and X-ray machines. Radiologists also needed large sources for teletherapy in treating deep tumors. We developed techniques for preparing a variety of ^{60}Co sources, ranging from tiny wires (to replace radon seeds) to 1000-curie sources for teletherapy. About the same time, researchers on radiation effects wanted large sources for irradiators. Since the radiographer commonly used "inches" when describing sources, whereas the teletherapist was more familiar with "centimeters," "building blocks" were made to satisfy all potential users: standard-sized pieces of cobalt were irradiated, including $\frac{1}{16}$- by $\frac{1}{16}$-in. to $\frac{1}{8}$- by $\frac{1}{8}$-in. right cylinders, 1- by 1-cm and 2-mm by 1-cm disks, and 2-cm by

1-mm wafers. The production of ^{60}Co was divided between Hanford and the MTR. Hanford reactors provided cobalt for radiography, and the MTR, material for teletherapy. The ever-increasing requirement for higher specific activity and the growing total demand for teletherapy material prompted us in 1956 to ask the AEC's Savannah River Plant to produce ^{60}Co for teletherapy sources; when these reactors were used, the logjam soon was broken. An interesting sidelight was the evolution of sealing techniques for encapsulated sources. The technology proceeded from single to double encapsulations with lead gaskets and screw threads and then from soldering to Heliarc welding. This evolution was intimately tied to improvement in remote-handling techniques in manipulator cells.

COBALT-60 STORAGE "GARDEN"

As a by-product of the ^{60}Co program, we completed in 1956 a storage facility for ready accessibility and dry storage of cobalt. Included in the design was a cavity for using the radiation emanating from the 100 thousand to 300 thousand curies of cobalt that were kept in inventory. The facility was named the Cobalt Storage Garden and Irradiator. The irradiator cavity provided 1 cu ft of dry irradiation space with a gamma dose rate in excess of 1×10^6 r/hr (the maximum dosage was dependent upon the configuration used in loading the storage tubes surrounding the chamber). Many materials have been irradiated in this cavity, but one we will not forget was the perfume brought to the Laboratory by a perfume expert. After irradiation at various dosages, the results, good or bad, were determined by the expert's smelling the samples.

FISSION PRODUCTS

Fission products were always a special category of radioisotopes—not "waste," but rather a bountiful supply of inexpensive radioactive and stable isotopes. Further, many of the nuclides that occur in fission are difficult to make in high specific activity by other neutron reactions. The short-lived fission products processed in the early days were ^{131}I, $^{95}Zr-^{95}Nb$, ^{140}Ba, ^{89}Sr, ^{141}Ce, ^{103}Ru, ^{91}Y, ^{147}Nd, and later ^{133}Xe. The feed for early work on these radioisotopes came from the effluent of an early classified program. Later, when this source of supply was no longer available, the irradiation of tellurium was considered for the specific production of the important isotope, ^{131}I. However, when it became apparent that meeting the demand for ^{131}I would require an inordinate amount of space in the Graphite Reactor, the decision was made to produce fission-product iodine by processing uranium "slugs." Now, from enriched-uranium targets irradiated in the ORR, this is still a source of high-specific-activity ^{131}I and other fission-product radioisotopes. The major effort and accomplishment in this program was the development of processes and relatively small-scale process units for separating and purifying these shorter lived materials. The work on longer lived fission products was concentrated on ^{137}Cs, ^{90}Sr, ^{106}Ru, ^{147}Pm, ^{144}Ce, and ^{99}Tc. In the early days a variety of processes were tried, including ion exchange. We remember well our initiation to the gassing problem when 1 curie of ^{90}Sr was placed upon a small column (~1 cm in diameter by 25 cm long). None of the solution added to the head tank flowed through the column because of the back pressure developed by gases formed through radiolysis! However, the development of appropriate processes led eventually to the large-scale separation and purification of these long-lived species. Some of the most difficult and exasperating experiences related to the procurement of appropriate feed materials, which were supplied from the ORNL Tank Farm, Idaho Chemical Processing Plant, and ultimately Hanford. The work was especially satisfying, however, because it culminated in the design and construction of the Fission Products Pilot Plant during 1954–1956. Few people believed that fission products would amount to anything, and appropriations for radioisotope capital equipment were always hard to get. Nevertheless, with the help of many people at various levels of company and government management, F3P (as it came to be called) was completed and operated with signal success, furnishing a fitting climax to the first decade of the radioisotope program.

In retrospect, these were probably the most exciting days of our lives. Even though many of them were trying, we always felt a sense of challenge and accomplishment while learning, doing, and making these interesting and useful materials available for use in such a variety of fields.

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Early History of Radioisotope Research and Development in the Chemistry Division at Oak Ridge

By G. E. Boyd

Dr. George E. Boyd joined the Manhattan District U. S. Engineers in 1942 as Section Chief, Analytical Chemistry Section, Plutonium Project, at Chicago and then transferred to the Clinton Laboratories at Oak Ridge, Tenn., in 1943. In 1948 he became Associate Director of the Chemistry Division, was Assistant Director of Research for the Chemical Sciences in 1954–1955, and has been Assistant Director of the Oak Ridge National Laboratory since 1955.

Dr. Boyd is currently Chairman of the Division of Nuclear Chemistry and Technology, American Chemical Society, and is on the Board of Directors of the American Nuclear Society (ANS); he is a member of the Committee on Ion Exchange Nomenclature, National Academy of Sciences, National Research Council, and ANS representative to its Division of Chemistry and Chemical Technology. He has held various journal editorships and is presently a member of the Honorary Editorial Board of *Radiokhimiya* (Radiochemistry) and of the Editorial Advisory Board of *Radiochemica Acta*.

His publications include many on pure and applied chemistry, physics and chemistry of surfaces, chemistry of technetium, and physical chemistry of organic ion exchangers.

The radioisotope effort was a gradual and logical outgrowth of the wartime efforts of the chemists and chemical engineers working at Oak Ridge on the Plutonium Project. The "proving-out" of the plutonium separations process in the Pilot Plant at the Clinton Laboratories in late 1944 allowed the attentions of both the fission-product radiochemistry and the plutonium separations process groups in the Chemistry Division to turn to problems on the preparation of carrier-free radioisotopes. Much of the radiochemical analytical work developed earlier for the separation and purification of the uranium fission products prior to their identification and characterization was adaptable to the isolation of these radioisotopes in much larger quantities. Some of the early work actually took place at the University of Chicago in 1943 following the creation of a "radiotoxicology" section of the Health Division; this section commenced the fractionation of the fission products with the aim of supplying pure carrier-free isotopes for project biological studies.

The chemists in the fission-product group at Oak Ridge (notably G. W. Parker, J. W. Ruch, and P. W. Lantz) also had experience in high-level radiochemical separations because of their successful efforts to separate kilocurie amounts of promethium for characterization and cross-section measurements, and of ^{135}Xe for poison spectrum measurements.

There was understandably a considerable interest by the chemists and others in developing the benign consequences of the release of nuclear energy, which doubtless was a reaction to the weapons aspects. The great importance of the availability of radioactive isotopes to science and to medicine was recognized, and the pre-1946 efforts by many persons too numerous to mention by name generated great devotion and enthusiasm. Most of this work was conducted on laboratory bench tops or in hoods with rudimentary shielding and semiremote manipulators. Later, in 1946 and 1947, after many of the chemists returned to universities to resume teaching or to work for graduate degrees, these operations were transferred to engineering and development groups where by 1948 a full-fledged radioisotope production activity was attained and multicurie quantities were processed routinely, in many cases using adaptations of the early basic chemical methods.

The potentialities of the Oak Ridge Graphite Reactor were realized quite early, and several lines of endeavor for the production, separation, purification,

and conversion of radioisotopes were initiated which continue to be active even down to the present time.

The formal organization of a radioisotope section in the Chemistry Division took place in early 1945. This section was led by W. E. Cohn, who served for approximately 1 year before being succeeded by J. A. Swartout, assisted by W. H. Baldwin and H. Kohn. Some of the members of Cohn's section were Parker, Lantz, W. R. Martin, G. E. Creek, S. W. Mayer, R. T. Overman, P. C. Tompkins, and E. R. Tompkins. Extensive experimental surveys were conducted in 1945 by Overman and later by Cohn on the preparation of many isotopes by neutron capture or transmutation reactions, which delineated the tremendous potentiality of the reactor as a production tool. As a result, the first "catalog" of those isotopes available from the Oak Ridge Reactor (MDDC-18) was prepared and subsequently provided the essential part of the June 14, 1946, announcement of the routine availability of isotopes.

During the period 1945–1946, members of the radioisotope section of the Chemistry Division developed and improved the production and handling methods to meet the potential public demand. Work was begun in 1945 on preparation of ^{14}C by L. Slotin and A. H. Snell, who were later assisted by L. Norris. For example, an ingenious loop was placed in the Graphite Reactor through which aqueous ammonium nitrate solution was circulated. The organic chemists in the Chemistry Division—including W. B. Leslie, O. K. Neville, L. Spector, and, later, C. J. Collins and others—aided in analyzing the products from this loop. The organic chemists also undertook an extensive program of research and development on the synthesis of ^{14}C-labeled compounds to make this all-important radioactive isotope more readily applicable for research in biology and particularly in medicine.

One of the important early achievements was the development of a process by Cohn for the separation of high-specific-activity ^{32}P from reactor-irradiated elemental sulfur. This product was for some time shipped to Berkeley for release as a cyclotron product, thus hiding not only its true origin but also the fact that the Berkeley cyclotron was irradiating uranium for the Oak Ridge chemists who were working on plutonium.

Work on the preparation of the medically important isotope ^{131}I was begun in 1946 by D. S. Ballantine using neutron-irradiated tellurium as a starting point. This was followed later by the work of Rupp on the preparation of fission-product ^{131}I.

Early work on the radiochemistry of technetium and the production and isolation of ^{99}Tc also derived from the wartime effort directed to the study of the fission products. Parker, Ruch, Boyd, E. E. Motta, Q. V. Larson, and others were some of the individuals connected with this effort.

Another line of effort that has persisted was the work on rare-earth separations with ion-exchange

Fig. 1 Pure technetium metal, 554 mg.

columns which grew out of the efforts beginning in early 1942 on the development of an ion-exchange plutonium separation process. Many of the lighter rare earths that occur in uranium fission could be isolated essentially carrier free using ion-exchange columns. Several groups were active in this work, and one of the by-products was the first separation and identification of the synthetic element promethium by J. A. Marinsky and L. E. Glendenin. Another by-product was the ultimate mass production of rare earths by Spedding et al. at Ames Laboratory. Other investigators working in this area in the Chemistry Division were E. R. Tompkins, J. X. Khym, P. C. Tompkins, C. D. Coryell, Cohn, B. H. Ketelle, L. S. Meyers, J. Schubert, and Boyd, several of whom had previously worked at the University of Chicago.

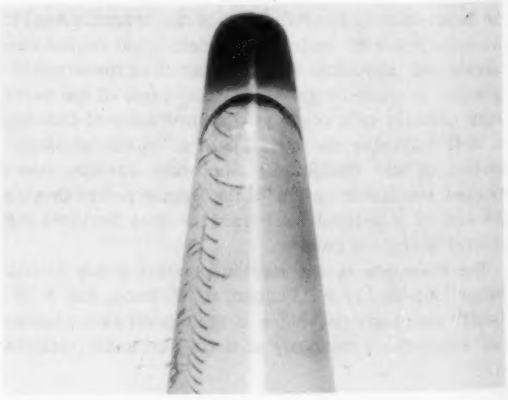
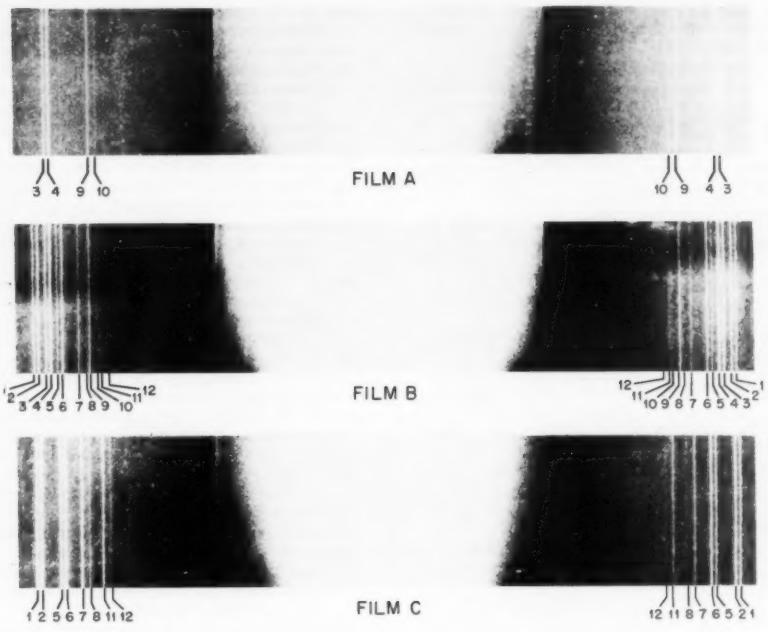


Fig. 2 Photomicrograph of promethium hydroxide.



Line identification

1 Nd K $\alpha 2$	5 Sm K $\alpha 2$	9 Pm K $\beta 1$
2 Nd K $\alpha 1$	6 Sm K $\alpha 1$	10 Pm K $\beta 2$
3 Pm K $\alpha 2$	7 Nd K $\beta 1$	11 Sm K $\beta 1$
4 Pm K $\alpha 1$	8 Nd K $\beta 2$	12 Sm K $\beta 2$

Fig. 3 K series X-ray spectra for promethium, neodymium, and samarium. Film A, promethium; Film B, neodymium, promethium, and samarium; Film C, neodymium and samarium.

A third line of investigation that has remained active was the preparation of high-specific-activity isotopes based on the use of the Szilard–Chalmers reaction in the chain-reacting pile. This work grew out of the researches by R. R. Williams, A. W. Adamson, and Boyd in 1944–1945, which was directed toward the preparation of high-specific-activity ^{124}Sb. Work was continued by S. Wexler and others who attempted to prepare high-specific-activity ^{59}Fe.

A great many ideas, in addition to those worked on, were rife during late 1944 and 1945. Some of them have not yet been followed up, although they appear as attractive today as they were 20 years ago. One of these ideas was the suggestion that radioisotopes could be prepared in extremely high specific activity by means of electromagnetic separations following irradiations in the Graphite Reactor. Although small-scale mass separators are operating today for the preparation of sources for nuclear spectroscopy studies, as yet no attempt has been made at "mass" production. The important possibility that chemical exchange reactions might also be used, especially to enrich low-mass isotopes of the periodic table, is as attractive today as when the idea was first broached.

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Radioisotope Characteristics, Measurements, and Standards

By S. A. Reynolds

S. A. Reynolds, Oak Ridge National Laboratory, came to Oak Ridge in 1944 as an analytical chemist with the Clinton Laboratories and in 1948 was named Leader of the Radiochemical Research and Development Group. From 1950 to 1958 he was an instructor for the Oak Ridge School of Reactor Technology. Since 1961 Mr. Reynolds has had special assignments in analytical chemistry and isotopes research and development. He is a member of the radioactivity measurements and standards subcommittee of the National Research Council.

When Oak Ridge National Laboratory (ORNL) began the radioisotope business, there were no generally accepted standards, and the knowledge of characteristics of the nuclides was very limited. Measuring techniques and standards had to be developed, and radioisotope characteristics such as half-lives and gamma intensities had to be accurately determined. This paper will survey the development of this field, and the knowledge and methods of today will be contrasted with those of 20 years ago. Because others[1–3] have reviewed the general progress from time to time, the present discussion is limited largely to ORNL work either done for the isotopes program or adapted to its needs, though major parallel developments elsewhere are covered.

Many people in the Analytical Chemistry, Chemistry, Instrumentation and Controls, Isotopes, Physics, and other ORNL divisions have participated in this work through active research, advice, or support. Some of the individual contributors who are still in the program are W. S. Lyon and E. I. Wyatt, Analytical Chemistry Division, and A. F. Rupp, Director of the Isotopes Development Center. Work of others is cited in the references. The U. S. Atomic Energy Commission (AEC) has made possible the whole program, particularly in recent years through its Division of Isotopes Development (DID); notable contributions were made by P. C. Aebersold, formerly Director of DID.

In each of the following subsections a brief historical summary is given, leading to the present "state of the art." Knowledge of isotope characteristics is inextricably involved with instruments and standards; so the division into separate topics, as done here for convenience, is necessarily arbitrary.

Decay Schemes and Half-lives

The decay scheme of a radionuclide is a description of its decay, giving the types and energies of the radiations emitted and including which gamma follows a particular beta or another gamma. Information on modes of decay, schemes, and methods of deducing them is given in a familiar text.[4] Twenty years ago the knowledge of some simple decay schemes was adequate, but complicated schemes were very poorly known and were usually thought to be simpler than they actually are. For example, ^{144}Ce was formerly believed to have no gamma,[5] while it is now known to have several.[6] Half-lives, especially those of a few months or greater, were also not well established. An example of the difficulties caused by this lack of knowledge is that, in the early days, the isotopic abundances of ^{14}C in the $BaCO_3$ products were calculated from the measured specific activities, using a conversion factor based on the then current half-life of 5100 years. The calculated isotopic abundances were therefore too low since, in a few years, the accepted value became 5568 years. E. I. Wyatt and the author issued an internal memo in 1953 containing local specific-activity and mass-spectrographic data that led to a half-life estimate of 5800 ± 100 years but declined to have it listed in tables because it was different from the accepted value. Our reticence is amusing in retrospect since the presently accepted "Cambridge average" is 5730 ± 40 years. Cesium-137 furnishes another example; in 1948, values for the half-life of 33 and 37 years were tabulated. Subsequently, published values dropped as low as 26 years, and the weighted mean is now

29.82 years, while the unweighted mean is 30.1 years. The author has been following the decay of four ^{137}Cs sources for 7 years, with the current tentative finding of 30.12 ± 0.21 years. Fluctuations of the apparent half-life with duration of measurements is quite striking: a plot is given in forthcoming Report ORNL-4039.

As time passed, physicists and nuclear chemists used more and more refined techniques to clarify the decay schemes and to determine the half-lives of radioisotopes; a significant portion of this work was done in the Analytical Chemistry Division.[7,8] Today the percentages of the decays of a given nuclide which go through various beta or positron branches (given in terms of the maximum particle energy, $E_{max.}$) or through electron capture (EC) are reasonably well known. The energies of the gammas associated with the various branches, or with isomeric transition, are also adequately known. The absolute percentages of unconverted gamma emission [γ photons/100 disintegrations or $100 \times (\gamma/\text{dis})$], however, are not sufficiently well known in many cases, and it is just these values which are most important in certain measurements and applications. These can be deduced from relative-intensity measurements — which are often available — if one knows the percentage of one gamma. Sometimes the percentage of a given gamma can be estimated from its conversion coefficient and the percentage of beta or EC populating its level. More such deductions by compilers would be useful in practical work.

SOURCES OF INFORMATION

The Nuclear Data Sheets[6] were prepared by the Nuclear Data Group headed by Dr. Katharine Way, formerly at the National Academy of Sciences and now at ORNL. This group's information will henceforth be given in the journal *Nuclear Data, Section B.* The contents and history of the *Sheets* and of the journal were recently reviewed.[9] For many nuclides particular values for constants such as half-lives, radiation energies, and radiation intensities are adopted; selection techniques have been described.[9] Some years ago a careful compilation of useful data for a number of common radionuclides was issued.[10] The National Research Council subcommittee on radioactivity standards is preparing a greatly enlarged version of this compilation, to be published as an issue of *Nuclear Data.* Other sources of information include *Nuclear Science Abstracts* (under index entries for particular nuclides), a well-known handbook,[11] and, of course, the many journals in the field. Data are often unevaluated, however; so one has little idea of accuracy.

SELECTION OF VALUES

Anyone who compiles or uses nuclear constants — for use in analytical measurements, shielding calculations, prediction of heat-source outputs, and the like — must often select values himself. Because of the large number of nuclides covered and the thoroughness with which each is studied, some of the Nuclear Data Group's published values necessarily lag several years behind current literature.

Great care must be taken in making choices, because the selection process itself is not necessarily clear-cut. The mean of published values for a given constant might be used, with each value weighted inversely as the square of the uncertainty ascribed to it. This is proper statistical procedure,[12] but experience has shown that it sometimes leads to badly biased estimates, because particular investigators have been overoptimistic about accuracy or have been unaware of some large systematic error. Further, some good values have had no error statement. The uncertainty of the mean is also weighted; so one value of very small apparent error can induce an unrealistically small estimate of this quantity. Another approach, also not universally satisfactory, is to take an unweighted mean of published values after rejecting outliers by some reasonable test[13] such as Chauvenet's criterion. This method suffers from the disadvantage that truly accurate values are not weighted as they should be. The author has sometimes used a compromise procedure, in which the mean is weighted but its standard error is computed from the unweighted differences of the individual values from the weighted mean.

The set of constants for each nuclide must be considered as a special case. For some nuclides, only one or a few determinations have been made, and, since no statistical approach can be applied, the scientist hopefully uses what he finds. In many cases he can take means and exercise some commonsense judgment. Generally, he should be suspicious if the apparently most precise value is very different from the mean of the others or if there is not general overlapping when the various uncertainties are considered. Plotting on probability paper[12] is sometimes useful; Fig. 1 is such a plot and illustrates some of the above principles. The points and error bars represent eight values compiled by the Nuclear Data Group, plus a recent determination by the author,[14] 8.070 ± 0.009 days. The weighted mean of all nine is 8.09, strongly influenced by the highest value, 8.141 ± 0.006, because of the small error ascribed to it. It is apparent that this value is an outlier. (In fact, it can be rejected by Chauvenet's criterion.) The median and both weighted and unweighted means of the remaining values are 8.06 days, and this is the recommended value.

The author and colleagues have selected decay-scheme constants (or determined them) for over 100 nuclides included in the ORNL radioisotopes program.[15-18] Most of these selections are identical with or close to those in *Nuclear Data Sheets* or forthcoming issues of *Nuclear Data.*

Instruments and Measurements

Twenty years ago instrumentation for the radioisotope program was essentially limited to G–M

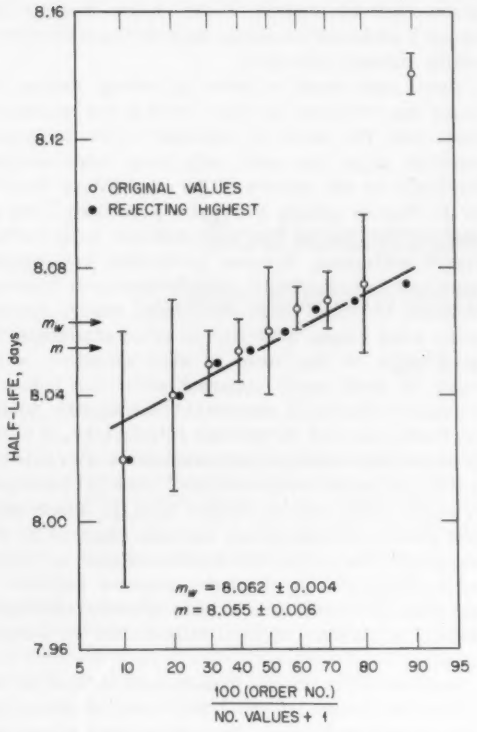

Fig. 1 Probability plot of values of half-life of ^{131}I.

counters for beta and gamma measurements and to proportional counters for alpha determinations. Zumwalt and coworkers were doing their pioneering work in measurement of disintegration rates of beta emitters.[2,19] Borkowski and associates were developing the high-pressure ionization chamber for gamma measurements and proportional counters for beta and X-ray (EC) determination.[20] Measurement of absorption in aluminum of beta radiation from a sample, with extrapolation to correct for air and counter-window absorption, was used often for "absolute beta counting."[2]

In the ensuing years great advances were made in instrumentation. A review of general developments will not be attempted here, because numerous contemporary reports[21-25] have been issued. Rather, a summary of the adoption of various types of instruments for routine analyses is given in Table 1, with appropriate references. Certain specialized techniques are listed in Table 2. A contemporary tabulation of ORNL methods was made some time ago,[40] and the calibration and routine measurement methods for 102 present and past isotope products have recently been summarized.[41] Detailed discussions of some of these are included in the ORNL Master Analytical Manual.[42] Recent general developments in instrumentation have been summarized.[43,44] The particular usefulness of the ionization chamber for high-energy gamma and of the scintillation devices for low-energy gamma is made clear by their energy–efficiency curves, Figs. 2 and 3. The American Society for Testing and Materials committees E-10, Radioactive Isotopes, and D-19, Industrial Water, are continually producing data on radioactivity.[45]

PRECISION AND ACCURACY

In nuclear measurements the familiar distinction between precision and accuracy must be made. Precision can be established for relative measurements, i.e., count rates and ionization-chamber readings, and quantities derived directly from these, such as a half-life or a material balance in a process. Accuracy, or probable bias, must be estimated for absolute quantities such as disintegration rates (number of curies), cross sections, and power outputs. Ideally, statistical techniques of determining precision might be applied to every step in the derivation of a quantity that has scientific significance. Then all the errors could be combined to arrive at the overall precision, which (in the absence of unknown error) would be equivalent to accuracy. Normally this approach is impossible, and an approximation must be made. If a standard of stated accuracy is available (see next subsection), the scientist can simply combine the observed or computed precision of his measurement with the stated accuracy of

Table 1 ROUTINE MEASUREMENT TECHNIQUES

Method or instrument	Year adopted	Typical uses	Reference
Gamma ionization chamber	1949	Measuring gamma emitters	2, 26
Beta ionization chamber	1951	$^{14}C(CO_2)$	27
Proportional-counter spectrometer	1952	^{55}Fe, ^{59}Ni	24
NaI(Tl) scintillation counter	1952	General, gamma emitters	22
NaI(Tl) spectrometer	1953	General, purity tests	28–30
2π beta proportional counter	1954	^{63}Ni, other beta	27
Alpha spectrometer (ion chamber)	1955	Purity tests	27
Multichannel analyzer	1958	General	30, 31
Liquid scintillation counter	1959	^{35}S, ^{14}C, ^{3}H	32
Silicon alpha spectrometer	1960	Purity tests	31
Anthracene beta spectrometer	1961	Purity tests	27
Ge(Li) gamma spectrometer	1964	General (high resolution)	33, 34

Table 2 SOME SPECIALIZED METHODS

Method or instrument	Application	Reference
Beta–gamma coincidence	Standardization of beta–gamma emitters	31
X–gamma coincidence	EC–gamma nuclides	24, 25
4π counting	Pure beta emitters	31
4π coincidence	Beta–gamma emitters	31
Sum-coincidence (gamma spectra)	Low-energy gamma (e.g., ^{129}I)	35
Absolute liquid scintillation	Low-energy beta	36
Ge(Li) diode	Decay schemes (high resolution)	33, 34
Computer resolution of spectra	Decay schemes and impurity determinations	37
Calorimeter	Large sources	38, 39

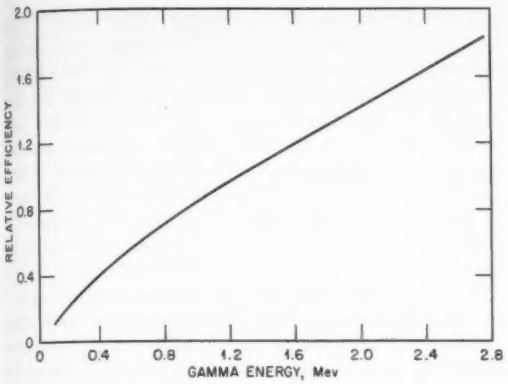

Fig. 2 Relative efficiency of 4π gamma ionization chamber.

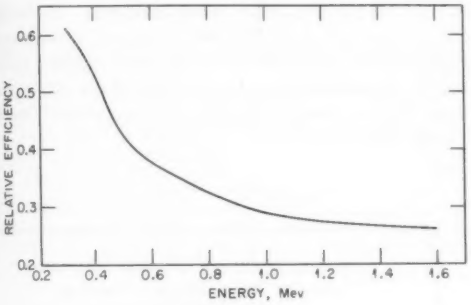

Fig. 3 Relative efficiency of well type scintillation devices.

the standard to obtain overall accuracy.[12] Generally, he should be suspicious of a quoted overall accuracy of less than about 1% for a measurement requiring disintegration-rate determination; 2 to 3% is usually more realistic.

An outstanding example of ignoring all but one source of error was given by a person (not of ORNL) who informed G. W. Leddicotte and the author how to determine the chlorine content of an organic material to 0.1% by activation analysis, during the early days of the use of that technique. His method was to form the material into a disk of convenient size, irradiate it in the Oak Ridge Graphite Reactor at known neutron flux for a carefully measured time, and count by means of a calibrated G–M counter long enough to accumulate 10^6 counts, for which the random error (ideally) is 0.1%. Therefore the accuracy of the chlorine determination would be to within 0.1%. Among the numerous uncertain quantities he forgot are the following: thermal flux and spectrum of neutrons, flux depression in disk,[46] cross section of ^{37}Cl, half-life of ^{38}Cl, efficiency for ^{38}Cl counting, self-absorption of beta in disk, etc., etc. A reasonable estimate of the error might be 20%—not 0.1%.

The subject of statistics of pulse counters is discussed in many books.[2,4,13,17,27,31] Precision of simple spectra from pulse-height analyzers can be estimated in the same way as count-rate data, but, if spectra must be resolved, the precision becomes poorer and the calculation so tedious as to require machine computation.[37,47] Artifacts that can cause errors in spectrometry have been discussed,[48] and an additional one is illustrated in Fig. 4, which is related to random summing, namely, creating a false peak at twice the energy of the real peak. This phenomenon is not as familiar as coincidence summing, and its occurrence can lead to errors in the analysis of isotope preparations to determine trace impurities emitting high-energy gammas.

Precision of level indicators (e.g., ionization chambers, rate meters) can be calculated, but it is usually more satisfactory to make a number of observations in the region of interest and calculate the standard deviation as usual.[12] Thus the author has found the precision of a typical gamma ionization chamber[42] to be within about 0.2% on scales where the background is less than about 1% of the sample reading.

A statistical criterion, *figure of merit*, has long been applied to selection of the best instrument for a particular application.[49] With the increasing availability of radioisotopes, largely through ORNL efforts, the best tracer–instrument combination can be selected; a simple treatment of the method for doing this has been given.[50]

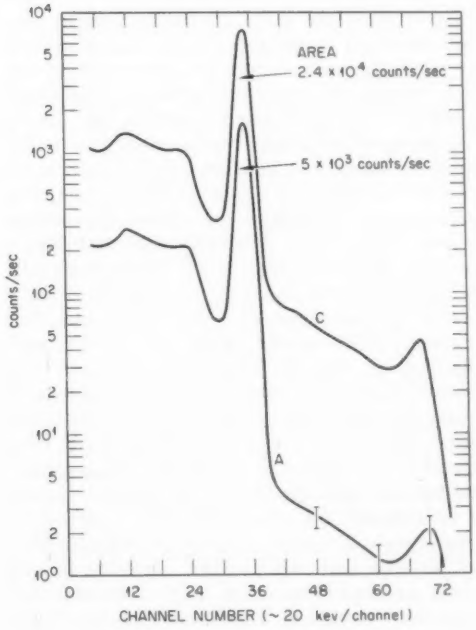

Fig. 4 Random-sum spectra of ^{137}Cs.

Determination of half-lives by direct decay requires relative measurements only; precision therefore needs to be considered. A weighted-least-squares computer program is useful for such calculations; CLSQ is a program[51] often used locally. Half-lives of many nuclides have been determined.[8,14] Obviously the presence of a contaminant can cause erroneous results. Because it shows the deviation of each point, CLSQ will sometimes indicate contamination or other systematic error through appearance of a trend; also, early and late portions of the decay study can be analyzed separately.

Radioactivity Standards

When a scientist makes a disintegration-rate measurement, he has — at least temporarily — a standard. He can employ precision calculations — and sometimes estimates of bias — to establish the accuracy of the standard.[52] Ordinarily a disintegration-rate standard (sometimes called, with dubious semantics, an "absolute" standard) is used to calibrate a measurement technique; at the same time readings are taken on a *reference source*, a long-lived emitter of appropriate radiation. Subsequent measurements of samples are normalized according to the response of the instrument to the reference source. Note that the *technique* is calibrated, not just the instrument. Conditions such as shape, positioning ("geometry"), and mass must be kept the same for standard and unknown, and if the instrument is nonlinear — having high dead time or gain shift, for example — standard and sample must be of about the same activity. In most cases a solution standard is desirable for ease in simulating sample measurement conditions. When energy–efficiency curves like the ones in Figs. 2 and 3 have been obtained for particular techniques, they may be used to produce "secondary" standards.[53] For work of ordinary accuracy, it is thus often unnecessary to use an "absolute" technique, and standards of short-lived material can be "maintained," i.e., new ones prepared, by measurements on a carefully calibrated device.

The first standards used in the radioisotope program consisted of U_3O_8 in a thin plastic or metal "sandwich" in the center of a rectangular plate that was inserted in the second "shelf" of a G–M counter (~2 cm from the window). Only the betas from ^{234m}Pa (UX_2) were counted, and such a standard indicated the approximately true "10% geometry" for energetic beta radiation. The need for standards of the nuclide to be measured — or a nuclide of very similar properties — quickly became apparent. Quite a few calibrations were made by Zumwalt and coworkers, as the author and associates were acquiring competence in standardization. Thereafter, for many years, calibrations were under the direction of the author, and E. I. Wyatt was — and remains — in charge of the Radioisotopes–Radiochemistry Laboratory.

While this work was in progress at ORNL, the National Bureau of Standards began to develop radioactivity standards. The first one (other than ^{226}Ra) was ^{131}I, which was distributed[54] in 1948. The results prompted the remark by G. G. Manov, "... we started 75 samples, got back 82 answers and nobody got the average."[23] The distribution curve of the results[54] is shown in Fig. 5. By the next year the iodine measurements had improved. During most of the history of the program at NBS, W. B. Mann has been in charge, and he has described the work.[23,25,53] In 1957 responsibility for the distribution of a number of standards was assumed by Nuclear-Chicago Corporation.

Very important in gathering information and sponsoring conferences was the Subcommittee on Measurements and Standards of Radioactivity of the National Research Council, chaired by Manov during most of its existence. Its successor is the NRC Subcommittee on Use of Radioactivity Standards, chaired by B. Kahn, U. S. Public Health Service.

Table 3 lists typical measurements of standards made at ORNL and other establishments. At ORNL these were treated as unknown samples, in approved quality-control fashion. Records show that for independent measurements of 83 preparations of 20 nuclides, over a period of 16 years, the overall mean deviation was 0.9%.

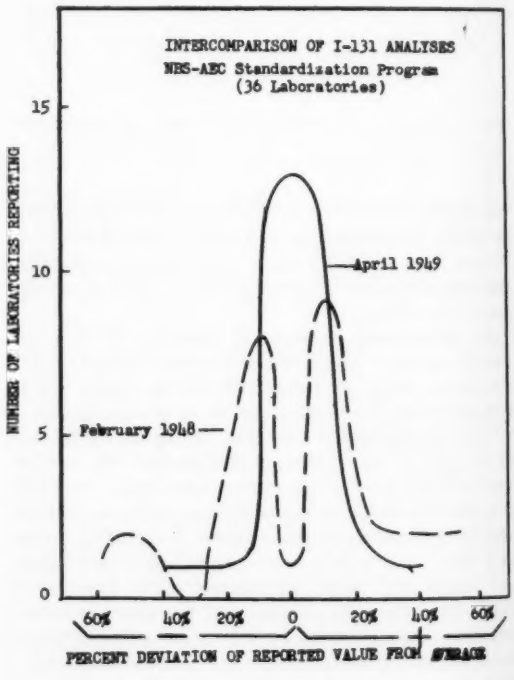

Fig. 5 Intercomparison of ^{131}I analyses [NBS–AEC standardization program (36 laboratories)]. This is a reproduction of the drawing No. 19171 as issued on Apr. 14, 1953; the quality of the graphic arts has improved tremendously since then.

Table 3 REPRESENTATIVE RESULTS ON STANDARDS

Nuclide	Supplier*	Year	ORNL half-life	Mean deviation, %
^{22}Na	IAEA	1963	2.60 years	1.0
^{24}Na	N-C	1960–1963	15.0 hr	0.7
^{32}P	NBS, N-C	1950–1963	14.3 days	0.7
^{35}S	N-C	1959	88 days	1.1
	IAEA	1962		0.0
^{36}Cl	NBS	1966	3.08×10^5 years	1.3
^{42}K	N-C	1960–1962	12.5 hr	1.2
^{55}Fe	NBS	1961	2.7 years	0.8
^{59}Fe	N-C	1959–1961	45 days	1.1
	IAEA	1962		1.0
^{57}Co	N-C	1966	267 days	0.6
^{60}Co	NBS	1953	5.26 years	0.0
	IAEA	1962–1963		0.1
^{82}Br	IAEA	1962	51 days	0.9
^{90}Sr	NBS	1957	28.6 years	1.6
	IAEA	1962–1964		1.2
^{90}Y	PHS	1961	64.4 hr	0.5
^{131}I	NBS, N-C	1951–1963	8.06 days	1.0
^{137}Cs	N-C	1960	30 years	1.5
	IAEA	1962–1963		0.6
^{144}Ce	IAEA	1962–1963	285 days	1.1
	NBS	1966		1.6
^{170}Tm	RCC	1966	127 days	0.9
^{182}Ta	N-C	1959	115 days	1.5
^{198}Au	NBS	1956	2.70 days	0.7
	N-C	1960–1963		0.8
	IAEA	1962–1963		1.1
^{204}Tl	NBS	1957	3.8 years	0.3

*N-C = Nuclear-Chicago Corp.; PHS = U. S. Public Health Service; NBS = National Bureau of Standards; RCC = Radiochemical Centre, Amersham; IAEA = International Atomic Energy Agency.

The present status of standards is about as indicated in a panel discussion[55] in 1963. Standards are listed as being available for about 60% of the 183 isotopes in a tabulation[56] by the International Atomic Energy Agency (IAEA). A number of these are "custom" calibrations, as provided at NBS. The Radiochemical Centre, Amersham, England, supplies solution standards for most of the radionuclides it produces, and IAEA currently distributes 20 solution standards and 8 solid gamma standards. National standardization programs have been described.[25,57] Two meetings on standards and measurements were held in the autumn of 1966. The conference on nuclear radiation measurement was held at Berkeley, England, September 12–16; and IAEA sponsored the symposium on the standardization of radionuclides October 10–14 in Vienna.

Future Program

Now that ORNL is no longer involved in the production of a great variety of radioisotopes, study of measurement methods, decay properties, and other data is expected to continue on a greatly increased scale. In particular cases, when an investigator working with an isotope has failed to find the information he needs, he might obtain it through the Isotopes Information Center.* In the short period of operation of the Center, several such inquiries have already been answered directly or forwarded to the special group having the information. Obtaining and measuring standards of various suppliers will continue. Collaboration with such organizations as ASTM, IAEA, and ASA will also continue.

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Early Developments in the Use of Radioisotopes in Agriculture

By C. L. Comar

Cyril L. Comar, Ph. D. in biochemistry, is Professor of Radiation Biology and Head of the Department of Physical Biology of the New York State Veterinary College at Cornell University, Ithaca, N. Y. After experience in several laboratories, including the University of Florida, he became director of the AEC Agricultural Research Program of the University of Tennessee, in Oak Ridge, in 1948. In 1954 he transferred to Oak Ridge Institute of Nuclear Studies as Chief of Biomedical Research, and in 1957 went to his present position. He is the author of numerous publications, including the book *Radioisotopes in Biology and Agriculture: Principles and Practice*, published by McGraw-Hill Book Company, Inc., in 1955.

This commentary is concerned primarily with the development of the use of radioisotopes in agriculture during the few years following the first shipment, in 1946, of reactor-produced radioisotopes under the auspices of the Manhattan District. It was the availability of reactor-produced radioisotopes and the associated technical advances that triggered the imagination and made possible meaningful applications to agriculture; and it was thus during the period from 1946 until the early 1950's that the principles, patterns, and techniques were established.

Although Hevesy had traced ^{212}Pb through bean plants as long ago as 1923, the early 1930's are regarded as marking the start of isotope usage in biological research. The very early studies were primarily of medical, physiological, or metabolic interest. Nevertheless, they were to serve as inspiration and guidance for the application of these procedures for agricultural purposes. For example, at the University of California, the work of Stout and Hoagland with radioactive phosphorus, potassium, and sodium in 1939 pointed the way to later soil-fertility research; the studies of Greenberg and colleagues with radioactive cobalt suggested a whole line of approach to problems of micronutrient deficiencies; and the work of Hamilton and others laid the basis for much that was to be done in mineral metabolism.

In 1935 Hevesy and colleagues reported on the use of radioactive phosphorus (^{32}P) for the first tracer experiment on the metabolism of this element in animals. Shortly thereafter the development of the cyclotron by Lawrence at the University of California made possible the production of varying amounts of radioactive isotopes of a few elements important in biology. In the late 1930's accelerators were built at a number of institutions such as the University of California, California Institute of Technology, Massachusetts Institute of Technology, the University of Rochester, and Cornell University. Biological and medical scientists working in proximity became aware of the existence of the radioisotopes and, with dabs of radioactivity left over by the physicists, were inspired to undertake a number of tracer experiments.

These early studies have been discussed in detail by Hevesy,[1] a great pioneer in the field. The efforts then were primarily to establish the metabolism of various elements in laboratory animals and plants. Most of the studies were done with radioactive phosphorus, particularly to determine its uptake by plants and absorption into the body with subsequent deposition in soft tissues and bone. Radioactive iodine was used to study the function of the thyroid gland, and radioactive sodium, to study the transport of this mineral across membranes into the various tissues of the body. Radioactive iron was used because of its relation to anemia of animals and chlorosis of plants. Biologists were keenly interested in the possibility of using a radioisotope of carbon because of the importance of this element as the backbone of essentially all important intermediary metabolites. The

cyclotron-produced radioisotope ^{11}C, despite its short half-life of 20 min, was effectively used to study the basic metabolic processes of both plants and animals. In early 1940 Ruben and Kamen demonstrated the production of ^{14}C, destined to become one of the most important radioisotopes in biological work.

In the early 1940's relatively few laboratories were using radioisotopes, and relatively few scientists in the biological field understood their use. By the early 1950's the use of radioactive isotopes had become widespread, and workers in practically every institution and laboratory had access to this technique to some degree. Between 1946 and 1952 more than 1000 shipments of radioisotopes for studies involving plants and animals were made from the production facilities of the Atomic Energy Commission at Oak Ridge, Tenn., to some 120 institutions located in practically every state, including Hawaii, and in Puerto Rico. In addition, specially synthesized radioactive compounds were available from commercial suppliers, and radioactive fertilizers were prepared and distributed by the U. S. Department of Agriculture (USDA).

The large-scale use of radioactive isotopes became possible because of vigorous concurrent development along three lines: (1) increased production of improved preparations of radioactive materials, (2) development of reliable instrumentation for the measurement of radioactivity, and (3) further education of scientists who were already competent in areas of biology and agriculture so that they could effectively apply the principles and practice of the use of radioactive materials.

Role of Radioisotopes in Agriculture

It was clear from the beginning that radioisotopes would not themselves be used directly to increase crop yield, but rather that the techniques would provide new tools for research and would aid agricultural production by improving and enhancing the efficiency of production methods. It may be of historical interest that there were serious claims of increased crop yields resulting from the direct use of radioactive materials; however, detailed experimentation did not support this view. It seemed that the application of radiation and radioisotopes would probably be manifested as the summation of many improvements, small and large, in plants and animals and in the technology of husbandry and processing as developed through research programs. These changes were expected to come about in a series of small steps, none of which would themselves be of tremendous impact but which ultimately were likely to be impressive in their total. A detailed report[2] from the Stanford Research Institute in 1959 attempted to estimate the probable economic benefits from the use of radioisotopes in agriculture and to analyze future applications on the basis of extensive surveys of work under way. In general, radioisotopes as a relatively new research tool made it possible to obtain certain types of information with more ease and accuracy and to gain new knowledge unobtainable in any other way. However, the overall benefits to agriculture may be classified generally as follows: (1) decrease of losses in production, storage, and distribution due to microbial action, diseases, insects, and weeds; (2) increase of productivity of land and other resources now in use through better understanding of the life processes, for example, development of optimal feeding and breeding; (3) development of new areas and resources, for example, through availability of power to provide irrigation facilities and to permit exploitation of farm resources. Specific uses to which radioisotopes were put, starting in the late 1940's are:

Soil and water
- Irrigation: study and measurement of water sources
- Soil composition, aeration, and structure
- Chemical analysis of soils for major and minor elements
- Organic decomposition
- Nutrient requirements and fertilizers: form, placement, uptake, leaching, sterilization, nitrogen fixation

Plants
- Uptake and loss of water and nutrients
- Movement within the plant of water, nutrients, and manufactured products
- Chemical composition
- Photosynthesis, respiration, and enzymatic processes
- Synthesis, degradation, and movement of proteins, fats, carbohydrates, vitamins, hormones, and growth substances
- Breeding of improved forms
- Reproduction
- Microbiology, including pathology and fermentation
- Environmental factors
- Action and residues of fungicides and insecticides
- Handling and storage

Animals
- Nutrition: management problems, requirements, deficiency symptoms, utilization of nutrients
- Physiology: lactation, renal function, liver function, gastrointestinal absorption, enzyme action, endocrine function
- Genetics and breeding
- Reproduction and fetal development
- Pathology: microorganisms, cancer, physiological disturbances, insects
- Metabolism: carbohydrates, proteins, fats, minerals

Entomology
- Insect dispersal and population studies
- Disease transmission
- Insect physiology
- Insect control: radiation, insecticides
- Longevity
- Parasitology

Veterinary medicine
- Diagnostic applications
- Therapeutic applications
- Disease processes: infectious, malignant, pathological, physiological
- Pharmacology

Engineering and mechanical processes
- Thermoradiation and temperature
- Time and efficiency studies
- Instrumentation

Start of Some of the Larger U. S. Agricultural Research Programs

Field experiments that could be adequately replicated with various crops were made possible by the availability in 1946 of ^{32}P in large quantities. Hendricks and Dean of the USDA, among others, took the lead in this work through cooperative arrangements with scientists from the Departments of Agronomy of the Agricultural Experiment Stations of North Carolina and Cornell University. Rapid expansion was made possible primarily through the development of a national cooperative research program involving soil and fertilizer investigations that had the support of several organizations, including the AEC, the USDA, and the fertilizer industry. Facilities were established at the Plant Industry Station, Beltsville, Md., for the production of radioisotope-tagged fertilizers. By 1952 ten different fertilizer materials had been produced for field experiments, including triple and ordinary superphosphates, and 94 field experiments were under way in many states. These experiments required about 35 curies of ^{32}P and involved some 18 crops. Some of the experimental objectives were the study of:

1. Comparative availability of fertilizer materials
2. Effect of granulation of fertilizers on availability
3. Effects of time and placement of fertilizer application
4. Effectiveness of surface applications to sod crops
5. Availability of materials applied as foliar sprays
6. Effect of lime, nitrogen, and potassium supply on phosphorus availability
7. Effect of irrigation on phosphorus availability
8. Comparison of crop species in utilization of phosphatic fertilizers
9. Residual value from phosphates in rotation and permanent fertility experiments
10. Availability of soil phosphates

In the subject area of plant physiology, Calvin and colleagues at the University of California used radioactive carbon to determine the numerous metabolic steps in photosynthesis, and Biddulph at Washington State College studied translocation in plants of radioactive iron and radioactive phosphorus as related to the causes of chlorosis. Arnold, Noggle, and Cohn in the Biology Division of the Oak Ridge National Laboratory also studied various aspects of photosynthesis, especially the biosynthesis of labeled sugars and intermediates and the effects of environment and radiation. Scully and Skok at the Argonne National Laboratory developed procedures for growing plants in the presence of ^{14}C and subsequently isolating from these plants labeled substances for use in later research. In this way ^{14}C-labeled sucrose, dextran, and various drug alkaloids were biosynthesized.

The use of radiation in plant breeding goes back to the mid-1920's when Muller and Stadler independently showed that X rays could increase the mutation rate in both animals and plants. Workers in Sweden then demonstrated the usefulness of this technique for practical plant breeding; Singleton, Konzak, and others at the Brookhaven National Laboratory developed a large radiation field with a ^{60}Co source in the center arranged so that plants could be grown in circles around it.

In the mid-1940's at the University of Florida, we were faced with problems of minor element deficiencies and, knowing of the work with radioactive cobalt that had been done at the University of California, started to use radioisotopes of cobalt, copper, and molybdenum for direct studies in domestic animals, especially cattle. The cobalt used in the early work was produced in the Massachusetts Institute of Technology cyclotron. After 1946 the studies were carried on entirely with radioisotopes obtained from nuclear reactors. In collaboration with others at the University of Florida, we also studied secretion of calcium into hen eggs and mineral availability to various crops.

In 1948, as a result of observations of damage to cattle exposed to radioactive fallout from the Alamogordo test, at Oak Ridge we developed, under the auspices of the AEC and the University of Tennessee, an extensive program based primarily on the use of a great variety of radioisotopes with domestic animals. The mission of this program was both to solve some of the immediate problems of radioactive fallout that were pressing and to develop an overall approach to the agricultural application of radioisotopes since the fundamental knowledge required for the latter was needed to provide a sound basis for the practical problems of food contamination. By 1952 Kleiber had well under way what he called the "Davis tracer team" for metabolic research at the University of California at Davis. His work was done with intact dairy cows and was based on the use of ^{32}P and ^{14}C for detailed studies of metabolism in the lactation process. Fundamental studies on lactation were also being started by Shaw of the University of Maryland and Peterson at the University of Minnesota. Also, about this time Wolterinck at Michigan State College was carrying on hormonal and nutritional studies with radioactive tracers in pigs and ewes, with special attention to thyroid function; and Clegg at Kansas State College, Jenkins at Fort Dietrick, and others were starting to use numerous radioisotopes in a great variety of ways to study the biology and physiology of insects, biosynthesis, disease transmission, insecticide toxicology, and control of and radiation effects on insects.

In 1943 Proctor and associates of the Massachusetts Institute of Technology initiated research on sterilization of foods by ionizing radiation. By 1950 interest in this process had grown, and research was being sponsored in a number of laboratories. Recognizing the great value to the armed forces of a commercially feasible process for radiation sterilization of foods, the Department of Defense undertook an extensive program of research in this field in 1953.

Some of the other institutions and workers concerned with various aspects were Brownell and colleagues at the University of Michigan, who worked on the design and facilities for the irradiation of food products, Johnson at the University of Illinois, who studied the effects of irradiation on the nutritive value of food constituents, and Kraybill and colleagues of the U. S. Army Medical Nutrition Laboratory, who initiated toxicological studies of foods that had been sterilized with ionizing radiation.

It has been possible, of course, to mention only a few of the individuals who were engaged in studies that had implications for agriculture at this time since numerous scientists were carrying on individual investigations in a great many different laboratories. The names and records of accomplishment are in the permanent literature.

Record of Progress

No useful purpose in this brief review would be served by any substantive discussion of the large numbers of the experiments and studies that were undertaken during the period of interest. The reader who desires this information can consult the published proceedings of the various conferences that have been held on the use of radioisotopes in agriculture. The first such conference, to my knowledge, took place at the Alabama Polytechnic Institute[3] in 1947. Emphasis was on bringing to agricultural scientists an awareness of the potentialities of this new tool.

Two conferences held in 1952, one at Oak Ridge[4] and the other at Kansas State College,[5] indicated an increasing number of workers in the field and a natural emphasis on the techniques that were becoming available for radioisotope usage. Results were reported on fertilizers, soil fertility, and plant nutrition; metabolic studies with farm animals; the use of radiotracers in studies of insects and insecticides; the important applications of ^{14}C in clarification of the photosynthetic process; and the beginnings of the production of radioactive products by biosynthesis for subsequent research.

Progress by all countries was evident at Geneva in 1955 at the First International Conference on the Peaceful Uses of Atomic Energy.[6] At a symposium in 1955 on Atomic Energy in Agriculture, sponsored by the American Association for the Advancement of Science,[7] several papers were presented on radiation sterilization of foods in addition to further developments in the fields of soils, plants, and animals. The Second International Conference on the Peaceful Uses of Atomic Energy at Geneva[8] in 1958 emphasized, among other matters, genetics and crop improvement by the use of radiation. At a conference entitled "Radioactive Isotopes in Agriculture," held in 1956 at Michigan State University,[9] emphasis was on herbicides and fungicides in plants and on waste-disposal problems in agricultural-research installations.

A conference in Mexico City in 1961, entitled "The Use of Radioisotopes in Animal Biology and Medical Sciences," dealt with subject matter restricted to the following major areas: general physiology, hematology, glandular functions, mineral metabolism, metabolism of lactation, and clinical studies.[10] A meeting on Radioisotopes in Animal Nutrition and Physiology in Prague[11] in 1964 stressed three specific subjects: the biochemistry and physiology of milk secretion, trace elements and magnesium, and the influence of environmental factors on the raising and reproduction of animals. The 1965 conference in Ankara[12] covered soil chemistry and physics, ion uptake and translocation, and fertilizer usage. The pattern seems clear: early meetings necessarily treated general uses of isotopes, but later ones were appropriately restricted to narrow subject areas. Logically as time goes on, it is expected that the organization of conferences will be governed more and more by the subject area rather than the use of any particular physical methods. Nevertheless, in the foreseeable future the concepts and techniques of radioisotope usage will continue in an important way to be superimposed on the fundamental disciplines such as biochemistry and physiology that underpin all agricultural research.

Trends

It is interesting to compare the work done 15 or 20 years ago with that being done today. Earlier studies usually involved ^{32}P and ^{131}I, which were important biologically and could be easily handled and measured. Then researchers began to use other radioisotopes of considerable biological importance but much more difficult to handle: ^{45}Ca, ^{35}S, ^{14}C, and ^{3}H. Today radioisotopes of over 80 different elements are available. In many cases two or more radioisotopes of the same element are being produced; this offers great advantages for studies that require repeated observations or corrections for secondary losses. Of particular interest from the agricultural standpoint are some recently available radioisotopes, for example, ^{47}Ca, which has the advantage that it can be easily measured by gamma counting without the need for sample preparation. Fluorine-18 and ^{28}Mg have recently made possible studies of these important elements which formerly could not have been done.

Earlier instrumentation consisted almost entirely of electroscopes or simple Geiger-Mueller counters. Today a dazzling array of equipment is available to those who have the necessary funds. Great advantages have been provided by crystals for scintillation measurements and especially by the techniques for liquid scintillation counting of those important biological radioisotopes ^{14}C and ^{3}H. The single-channel gamma-ray spectrometer and more recently the multichannel analyzer have made possible studies involving the use of several radioisotopes simultaneously. This is

especially important in agricultural work since often a great deal of time and expense are involved in obtaining the experimental plants and animals in proper condition for an experiment.

Great advances have been made in autoradiography. Formerly, only gross autoradiography could be done; now we can visualize deposition in parts of cells, such as chromosomes and mitochondria. This has come about by advances in both light autoradiography and a combination of electron microscopy and autoradiography. Use of the whole-body counter has made it possible to measure retentions in domestic animals by direct counting rather than by collecting excretions and subtracting amounts excreted from intake. Other important techniques in routine use include gas-phase counting, paper chromatography, electrophoresis, ion exchange, and neutron activation.

Conceptually, early studies were devoted primarily to determining simple isotope distribution. Today emphasis is on the resolution of complex metabolic cycles, for instance, the carbohydrate cycle in lactating animals and in photosynthesis. In the area of protein synthesis and transfer of genetic material, many ingenious experiments have been devised; for example, it has been shown that, after virus deoxyribonucleic acid enters bacterial cells, it makes its own messenger ribonucleic acid (RNA), which then attaches to the bacterial ribosomes to make new protein under its direction; by pulse labeling, it has been shown that the life of a messenger RNA molecule is about 2 min and that each messenger makes 10 to 20 molecules of proteins; it has been determined that protein is made by sequential addition of amino acids to a steadily growing chain; and the code is being unscrambled by isotope determination of nearest neighbor frequencies.

In early work in animal nutrition, simple balance tests were usually done to measure retention as an indication of absorption. More recently emphasis has been given to measurement, by use of radioisotopes, of fecal endogenous losses, and thus more meaningful values can be derived for the net absorption of elements and maintenance requirements. By measuring body composition at two time intervals, we can determine any change in body content that has occurred; this is much more accurate than measuring intakes and excretions and thereby the balance over a given time period. In earlier years simple isotope dilution studies were done, for example, determination of plasma and blood volumes. It is now possible to use multiple counting techniques. For instance, it is feasible in a single study, by simultaneous administration of radioactive sodium, potassium, bromine, and tritium, to determine (1) exchangeable sodium, potassium, and chlorine in the body; (2) plasma sodium, potassium, and chlorine; (3) total body water; (4) extracellular water, sodium, and potassium; and (5) intracellular water and potassium. In earlier years simple diagnostic methods were used, such as simple thyroid function tests and iodine uptakes. Today there is a possibility of developing many diagnostic methods, including liver and kidney function tests and determination of rates of spermatogenesis and transport.

Recently more advanced research is being conceptually concerned with kinetics of movement of substances between compartments and pools, with elucidation of steps involved in transport during intermediary metabolism, and, finally, with clarification of mechanism and function in biological systems.

There is also increasing emphasis on direct application of the response of biological systems to ionizing radiation. This approach is needed for procedures that deal with production of beneficial mutations for plant breeding, for sterilization of food, and for insect control. This type of study requires coordination and knowledge in the fields of genetics, reproduction life cycles, and physiological response to radiation.

We have come a long way from the glamour and excitement of the early days when one could quote as the standard definition of the radioisotope: "A substance which, when administered to a biological system, produces a scientific paper." At present it is fair to say that no important obstacle exists to the widespread use of nuclear energy techniques in biological and agricultural research. Training centers and courses are available; necessary equipment is obtainable; a broad variety of radionuclides is available; and many substances of biochemical interest can be obtained in labeled form. For these luxuries we must thank the physical scientists; but the challenge and responsibilities are in the hands of the biologists and agriculturists, who must learn to put these techniques to work meaningfully for their own purposes.

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Radioisotopes in Medicine Immediately After the Great 1946 Deliverance

By Marshall Brucer, M.D.

Marshall Brucer, M.D., was Chairman of the Medical Division of the Oak Ridge Institute of Nuclear Studies from December 1948 until 1962. Multiple sclerosis forced upon him an early retirement in 1962. But this retirement has been only partial. Now living in Tucson, Ariz., he is not only actively engaged with work in an isotope laboratory there, but is also an editor of the *International Journal of Applied Radiation and Isotopes* and the writer of "Vignettes in Nuclear Medicine," which is published by Nuclear Consultants Division of Mallinckrodt Chemical Works, St. Louis, Missouri. The titles alone of his other signed publications would fill pages. He is not a newcomer to *Isotopes and Radiation Technology*, having written the feature article of Vol. 3, No. 1, "118 Medical Radioisotope Cows."

It is painful to think back to the great events that occurred in nuclear medicine during 1946 because the great events weren't very great; in fact, it is hard to dredge up a few things that could be called events. A large segment of the physician population was busy getting out of the Army, and those who hadn't been in the Army were doing exactly what they had been doing all along.

To physicians who lived beyond Madison Avenue, Oak Ridge gave out only a minor blurp in August 1946. Nuclear medicine had to grow for about 10 years, and it took a lot of flaunting of their one great success, "The Atomic Cocktail," before many physicians, let alone the public, were aware that something had happened. To be sure, the first shipment of the Oak Ridge isotopes went to a hospital, but, beyond the address on the shipping ticket, this was too early in the game to have much to do with medicine. The Oak Ridge National Laboratory (ORNL) undoubtedly has a ream of records on the really first, first shipment, but this wasn't nearly so important as the publicitywise first "commercial" shipment which, except for the historical necessity of having an official first, could have gone simultaneously to perhaps six different hospitals. For those of you whose geometry stops at three dimensions, six simultaneous "firsts" might not make sense. But publicity, even in 1946, was omnidimensional; six simultaneous firsts was relatively modest. Today, the first shipment would have to go to about 5000 different hospitals, and there wouldn't be any necessity for the publicity ploy.

Rather than review the literature (rather sterile during this period), I will stick fairly close to what happened in Oak Ridge, because I lived through it. It wasn't too different from what happened throughout medicine. Most of us working with radioisotopes knew each other, and any success that one had was immediately copied by the others. We were first in some things, and others tried to grab the credit. Others were first in most things, and we made little effort to keep from getting the credit. None of us, incidentally, accepted the blame for anything.

In 1946 Priority in Radioisotope Development and Applications Was a Matter of Journalism

A significant danger in 1946 was "international control of the atom." The working politicians didn't know that the English, and the French, and the Italians, and the Germans, and the Russians, ... and the Japanese had a pretty good idea of what the atom was all about. We in America felt that we had a complete monopoly on the secret of the ages, and we were offering it magnanimously to the entire world. Amidst all the bickering about how much to reveal, the only thing we could agree upon was that atomic energy cured cancer. Therefore Congress gave the still-jelling first Atomic Energy Commission (AEC) [permanent peacetime civilian organization activated on Jan. 1, 1947, to succeed the Manhattan Engineer District (MED)], its first job (after the first first job

of making bombs, of course). Radioisotopes were to be made available to the basic sciences and for medical therapy.

Even though medical therapy had the first priority, the basic sciences had a somewhat firster first, and this was acceptable because we all knew instinctively that once you knew the cause of a disease the cure would be discovered forthwith. Of the three most important causes of death during our lifetime, see how basic science has triumphed. The cause of tuberculosis was discovered in the 1870's, and we all know how quickly the cure was then found. The cause of malaria was discovered in the 1890's; the current U.N. flap is just a mosquito-control project. The cause of schistosomiasis was known as early as 1906. And although schistosomes kill practically no one in the United States (and neither does the dreaded hippopotamus bite, for that matter), they are still the cause of many fatalities and much human suffering in Asia, Africa, and South America. In 1946 the thing we needed most was a crash program to make the radioisotope cure for cancer available to all humanity.

To implement the desires of Congress, cancer-wise, the first AEC, in addition to shipping out the ^{14}C cancer cure, set up a "single" Medical Division for the study of the therapy of cancer and allied diseases.[1] One of the single Medical Divisions was to be at the University of Chicago because that's where the first reactor was built. Another of the single Medical Divisions was set up at Brookhaven. A third Medical Division was set up at Oak Ridge because that is where the isotopes came from, whether reactor produced or from processed cyclotron targets. (You simply must forget about Hevesy and the Danish nuclear production of ^{32}P and the California cyclotron and all the work in Boston with radioactivity. Except for a few isolated cases, the cyclotrons were unavailable; the other reactors were still pretty secret; and, if you mention Boston, you have to mention Chicago, and San Francisco, and St. Louis, and Portland. There's no end to the complications. If you want this story to be a smooth narrative, logically progressing from myth to myth, you will have to forget these historical irrelevancies.)

For the Oak Ridge cancer cure, the MED initiated talk in 1946; the AEC continued talk in committee during 1947 and made the decisions in 1948. In 1949 the medical program at Oak Ridge was already started, and it was really aimed at the cancer therapy that Congress had envisioned. Among the promising candidates for cancer therapy were such isotopes as ^{72}Ga, ^{198}Au, ^{131}I, and ^{60}Co.

The First Big Failure Was Quite Informative

Gallium-72 had a 14 1/4-hr half-life. (Don't say this is wrong for 14.25 hr was a pretty good measurement in 1948, and it was certainly better physics than it was medicine.) One of the criteria for a Medical Division in Oak Ridge was that short-lived isotopes couldn't be studied far from a reactor. (Remember, this was 1949.) The cancer for which gallium was a specific was not being successfully treated by other means, which was another one of the criteria. Special equipment was needed for producing the radioactive drug and for measuring it, both chemically and radioactively. The "safe use" had to be investigated. Almost everything about the use of ^{72}Ga had to be developed from scratch.[2] (Remember, this was before pharmaceuticals, the medical spectrometer, or the Great Radiation Hysteria.)

My oral report on ^{72}Ga to the Radiological Society of North America was "Gallium stinks." (Don't bother to look this up in the literature because I didn't put it so neatly and concisely in writing.) But gallium stunk only insofar as the patient was concerned. We relearned that, insofar as the internal use of radioisotopes was concerned, chemistry took priority over physics. All of us relearned that radiation was not necessarily the "answer to cancer" (a phrase from the 1920's). The physicians involved relearned that radiation was "a two-edged sword" (a phrase from the 1900's).

The intracavitary use of ^{198}Au (Ref. 3) was a major study in the early 1950's. The radiation didn't cure any cancer, but it did cure one very debilitating symptom (intractable fluid accumulation) in a few cancers. Through this story the physicians involved re-relearned that the chemistry of a radioisotope is not so all-fired important after all, that radiation is at least a partial answer to cancer, and that, even if radiation is a two-edged sword, the safety edge is awfully blunt.

We all knew for sure, in the early 1950's, that the success of ^{131}I (Ref. 4) was due to the iodine specificity for one kind of tissue, and so we looked for other specificities for other kinds of tissue. Hafnium[5] was found to be specific for the adrenal cortex. Nothing like iodine, of course, but this was the day of the Differential-Absorption-Ratio. The ratio was a number, and a bigger number was a bigger number, no matter how you looked at it. We found a high concentration of sulfur in some tumors; this didn't do any good therapeutically, but there was definitely a bigger number. We found that the trivalent organic complexes of antimony had a higher concentration in red cells and liver, and the pentavalent complexes ought to be even better. The hematoporphyrins were found to be capable of carrying many radioactive metals. And so we re-re-relearned that chemistry had a place after all.

Interspersed Among the Failures Was an Occasional Success

But we did have some early successes, and they were in "therapy" (they were also in "cancer"). While sitting in a bomb shelter during the London blitz,

Leonard Grimmett was reading some accumulated physics journals to take his mind off the bombs. He read about a new isotope of cobalt with radiation characteristics that reminded him of the difficulties he had in developing the "radium bomb." The radium bombs were having considerable success in treating some cancers, but there was never enough radium to make the bombs truly practical. With ^{60}Co he could double the effective dose rate; in fact, he could double the loading of the instrument to an enormous 10 curies and quadruple the effective radiation dose rate. When the Medical Division was still being organized, ^{60}Co was already being used as a "substitute for radium" in brachytherapy. There was no reason why it couldn't also be a substitute for radium in teletherapy.[6] On a consultant visit to Oak Ridge in the abandoned E wing of the wartime Oak Ridge Hospital, Grimmett and I met with Paul Aebersold, then Head of the Isotopes Division of AEC, to broach the delicate question of using an unheard of 10 curies of ^{60}Co all in one piece.

"Wadaya wanna stop with 10 for?" said Aebersold. "I gotta coupla hundred cooking in the pile." Not knowing any better, I pushed it up to a thousand. Grimmett chose to ignore the production problem and went home to design a 1000-curie teletherapy machine.[7] Aebersold probably realized that he was obsoleting his somewhat unauthorized stockpile of 10-curie ^{60}Co slugs. All the mass of budgetary justification for the ridiculous idea of a 1000-curie source came much later.

In a few years, when the teletherapy machine was being introduced to the manufacturers of therapeutic X-ray equipment, a poll was taken of how popular the "new" supervoltage ^{60}Co would be. One highly respected commercial consultant didn't think the idea very good and predicted that in 10 years there would be only 30 machines in use; the most optimistic manufacturer predicted 300. I was not responsible to any auditor for my advice, and so, as a joke, I predicted 1000. By the time a decade had passed, well over 1700 ^{60}Co teletherapy machines were in operation. But then, nobody was keeping count any more. Still a few years later, a symposium was held at a radiology meeting on the subject of whether the ^{60}Co machine made the 250-kv X-ray machine obsolete. The answer was, "Obviously no!" In this same sense the Model T Ford is not obsolete either; they're worth more each year.

The Medical Significance of Radioisotopes Is Not Necessarily a Medical Subject

The first problem the Medical Division took up was "fume hoods" — a machine problem, not medical. Fortunately, ORNL took over the development. The fact that we even saw a problem is illustrative of how similar we were to the rest of the medical buttinskies. Fume hoods were not a medical problem to begin with. Medical radioisotope scanning, a UCLA invention (out of England, sired by Cassen), was a continuing problem, but it is not limited to cancer and certainly is not therapeutic. None of the external counting or detection devices are so limited.

We had no responsibility for what was probably the biggest success of the Medical Division (even though we did get a kind of glory-by-association from it). The Bell–Francis–Harris medical spectrometer,[8] an ORNL development, would justify our existence (and that of the National Laboratory) if nothing else had been done. None of the external counters as used in clinical medicine (and little of the therapy) could have had any quantitative value without the development of the medical spectrometer. It doesn't take much quantitation to make most clinical tests truly quantitative, but it does take some. The medical spectrometer wasn't the last word in physical precision, but it was more than adequate for clinical purposes. None of the external detection equipment that grew out of the first 10 years following the release of radioisotopes to the medical public was directed specifically to cancer, and certainly little was necessary in radioisotope therapy. But cancer therapy was what Congress was thinking about in that first Atomic Energy Act of 1946. But then, neither was the first shipment of a radioisotope from Oak Ridge even remotely involved in cancer therapy.

The truly new development of nuclear medicine, though not *caused by*, is probably best *symbolized by* that first shipment of radioisotopes in August 1946. You can't point to a single great medical idea that starts in August 1946. There hasn't been a single breakthrough, not even a crackthrough, that grows out of the first shipment. But something far more important did occur as a result of 10 years of trial and error. A fairly large segment of physicians learned from radioisotopes that sometimes chemistry is important in clinical medicine, sometimes it isn't. We also learned that sometimes physics is important in clinical medicine, sometimes it isn't. These in themselves are astounding developments.

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Early Use of Radioisotopes as Tracers in the Petroleum Industry

By Bernard A. Fries

Dr. Bernard A. Fries is a research chemist at Standard Oil of California's Chevron Research Corp. (formerly California Research Corp.) in Richmond, Calif. He was a research chemist with the Manhattan Project in its earliest days, first at the University of Chicago, then at Oak Ridge, and finally at Hanford. In 1945 he went to his present affiliation, where he has been active in developing tracer uses for radioisotopes in the petroleum industry, particularly in engineering applications. A biochemist and physiologist by graduate training, he is especially well qualified to assess the safe use of radioisotopes.

The use of radioisotopes for industrial tracing purposes is commonplace today, but in the mid-1940's it was a novel idea. The petroleum industry began to take advantage of these new tools quite early and, in the 10 years following the first shipment of reactor-produced radioisotopes, developed several procedures that are still in continuous use.[1] These early procedures included tracing of liquids, for example, in pipelines and process vessels, and tracing of solids, for example, abraded material from an engine part to determine wear of the part and thus evaluate the lubricant used.

Tracing of Liquids

PIPELINES[2]

The petroleum industry transports products of many different types through the same pipeline, identifying the boundaries between two adjacent hydrocarbons so that each may be distributed to the proper vessel upon arrival at the refinery or storage point. Common methods of making this identification are colorimetric determination of dyes included in the products or determination of the oil density at intervals. Both methods are tedious and time-consuming and require samples to be taken regularly or a portion of the stream to be bypassed. In 1947, less than a year after the first shipment of artificial radioisotopes, we began to investigate the use of ^{140}Ba–^{140}La to follow this interface. Our initial efforts were successful, since we found we could follow a ^{140}Ba-labeled interface instantaneously by means of a Geiger counter located outside the pipe wall (Fig. 1). The method was thus much simpler than the earlier methods, and it turned out to be more precise and sensitive, since detection of the interface occurred several minutes earlier (Fig. 2) owing to the time lag in filling the gravitometer. Also, the new method was not expensive—the 1 mc of radiobarium needed cost less than $3 then and can be bought for even less now.

Since the tracer must (1) have a half-life at least comparable with the time it spends in this line, (2)

Fig. 1 One of our associates, Dr. Donald E. Hull, using a Geiger counter to detect the passage of radioactive tracer injected into an oil pipeline during research on the radiotracer interface-marking technique.

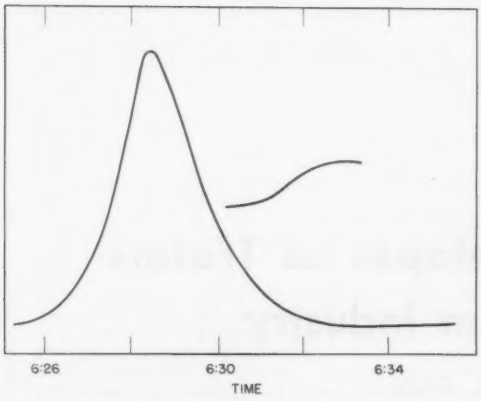

Fig. 2 Time of arrival of oil interface at a pipeline station as shown by the radioisotope tracer and density methods. The radioisotope method signals the interface approach 4.5 min earlier than the density method, and the sharp peak of the radioisotope wave indicates the interface arrival 3 min earlier than the midpoint of the broad density wave.

emit a gamma radiation that can be detected outside the pipe, and (3) be available as a stable, oil-soluble compound, ^{140}Ba proved to be a good isotope for this use. It has a 12.8-day half-life, its ^{140}La daughter emits a 1.6-Mev gamma radiation, and it is available as a chemically stable, oil-soluble soap. We also found ^{124}Sb, with a 60-day half-life and a 1.7-Mev gamma radiation, to be even more suitable in the form of triphenylstibine.

We tested the radioisotope tracer method on two short pipelines, one 5 and the other 24 miles long, and then made full-scale tests on the 182-mile crude pipeline from Rangely, Colo., to Salt Lake City. All results were favorable, and we finally put the method into use on a 560-mile line. In this operation we demonstrated considerable economies by its use. For example, in a long pipeline the interface becomes blurred by mixing of the leading and trailing stocks, and the mixture must be processed separately from either of the two stocks. Naturally the sharpness with which the widened interface can be determined becomes an important factor. When the radiotracer method was used, the usual 500 bbl of mixture was decreased 30% in the 560-mile line, with attendant decrease in re-refining time. In addition, station operators along the line were saved the time usually spent in watching with a gravitometer for the interface to pass by. No special radiological safety precautions were necessary once the isotope had been injected into the interface, and upon arrival at the terminal, the remaining isotope was diluted sufficiently that personnel hazard was less than that from cosmic rays. The process was reliable, and only normal maintenance was required. The method was finally patented and is now in routine use in a number of transportation lines.

IN THE FIELD

In another application the acidizing of a well was followed with radioisotope-labeled acid. The acid was pumped into a well, and its exact location in the well was observed with a Geiger counter.[3] In this way it was possible to be certain that the proper formation was treated.

REFINERY PROCESSES[4]

One of the earliest reported uses of radioactivity in the refinery was in the study of distillation processes. For determining the flow characteristics of a bubble-cap-plate vacuum still, ^{124}Sb, as triphenylstibine, was injected through the heavy reflux line into the center of the column (Fig. 3). Counting data obtained at various places on the outer wall of the column showed a nonuniform liquid flow, which was subsequently corrected to improve operation. Preliminary tests to determine whether a flow pattern can be found with a radioisotope were made with the only suitable tracer on hand at that time—5.27-year ^{60}Co as cobalt naphthenate, However, for full-scale tests more tracer was required than would be convenient with a 5-year-half-life material, and we therefore made arrangements with Oak Ridge National Laboratory (ORNL) to obtain the 60-day ^{124}Sb that was finally used in the larger scale tests.

In another vacuum still, entrainment in the vapor was followed by testing the distillate for ^{140}Ba–^{140}La, which was injected at various places in the stream.

The reason for an abnormal loss of catalyst in a "cat cracker" was found by using catalyst beads in which a radioisotope (^{95}Zr) was incorporated. The results of the test indicated that the loss of catalyst was due merely to a too rapid circulation rate, and the trouble was readily corrected. The test, which was made with a minimum of effort, averted a $100,000 plant shutdown, and the method was quickly installed in other plants as a routine control procedure.

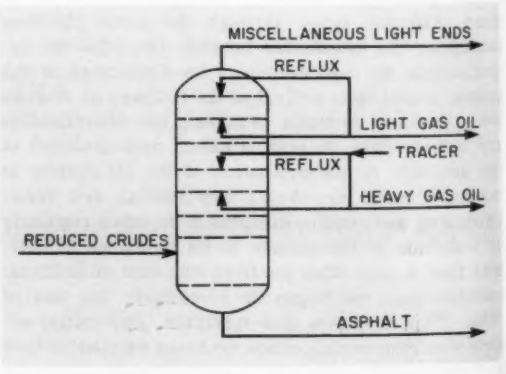

Fig. 3 Simplified diagram of bubble-cap-plate vacuum still.

Other problems in the refinery also yielded to the new method. Insufficient mixing in a surge tank that fed a cracker was demonstrated with the ^{95}Zr chelate of thenoyltrifluoroacetone, and the process was subsequently modified to produce a more homogeneous tank effluent. In a case where segregation of the product from a special batch of crude oil was necessary, ^{60}Co naphthenate was added to the still feed at the moment of switching from the regular to the special crude. This nonvolatile compound, which is soluble and stable in hot oil streams, follows the tar; when the tracer peak was observed in the bottoms effluent, the tar stream was switched to a different tank. Tests on the amount of radioactivity remaining in the product showed the method to be safe.

Wear Tests

Radioisotopes were early found to be useful in determining wear of machine parts.[5] A method was developed in which a cast-iron piston ring, for example, was irradiated with neutrons from the ORNL Graphite Reactor and installed in an engine. Ring wear under operating conditions was then determined by measuring the radioactive iron released into the lubricating oil. As little as 1 part in 10^7 of iron in the oil can be detected. The method has obvious advantages over the slow and expensive method of making physical measurements on engine parts before and after an extended period of operation, and it can show transitory effects, which the more cumbersome method cannot. It is also quicker and more sensitive than the method in which lubricating oil is chemically analyzed for iron. Wear rates determined by the use of radioisotopes correlated well with field data.

Significance of Radioisotopes

In a review[6] on the use of radioisotope tracers in refinery operations, King, just 11 years after the initial investigations, listed their use in the study of liquid, fluidized-solid, and gas flow rates; catalyst behavior, including mixing, inventory, and stack losses; entrainment; process mixing; reaction mechanisms; and leaks. His idea of the importance of these materials is shown by his own words:

> Radioisotopes are important tools in the progressive operator's kit. Future generations will wonder how a refinery was ever run economically without them. Uses are increasing more than exponentially, and oil companies measure savings in millions of dollars per year.

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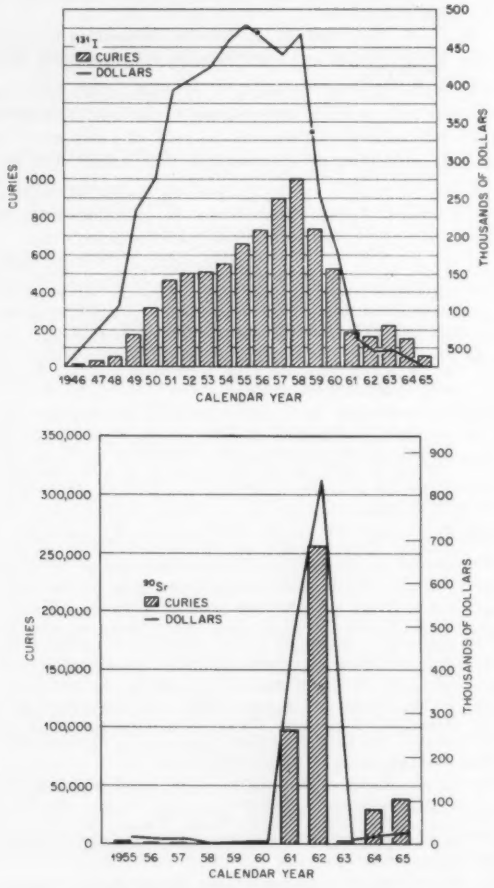
Twenty Years of Radioisotopes

The following graphs pretty well tell the story of the radioisotopes program over the past 20 years at Oak Ridge National Laboratory (ORNL). The data for the first 15 years are representative of the United States as a whole because ORNL was by far the major producer of isotopes in this country. Since then, as private U. S. suppliers and other countries have taken over the production of certain isotopes and ORNL has withdrawn in keeping with the policy adopted by the AEC,* the makeup of the figures has changed, but the overall picture has continued to show a steady growth pattern. The figures themselves are self-explanatory.

*See *Federal Register* for Mar. 9, 1965: Policies and Procedures for Transfer of Commercial Radioisotope Production and Distribution to Private Industry.



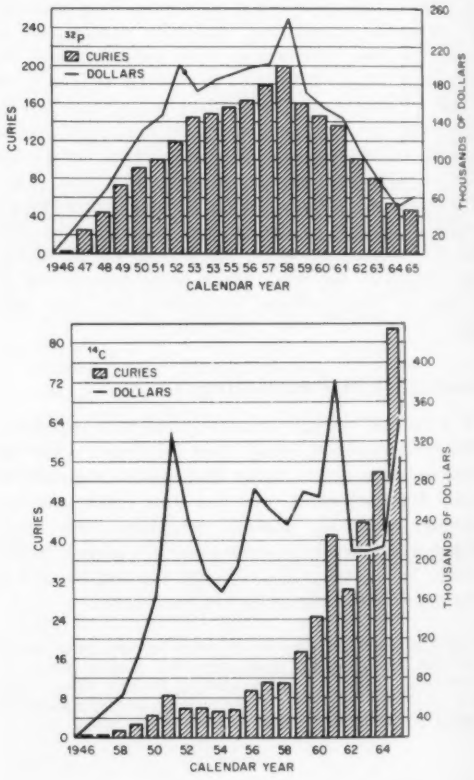


Fig. 1 Distribution of typical radioisotopes.

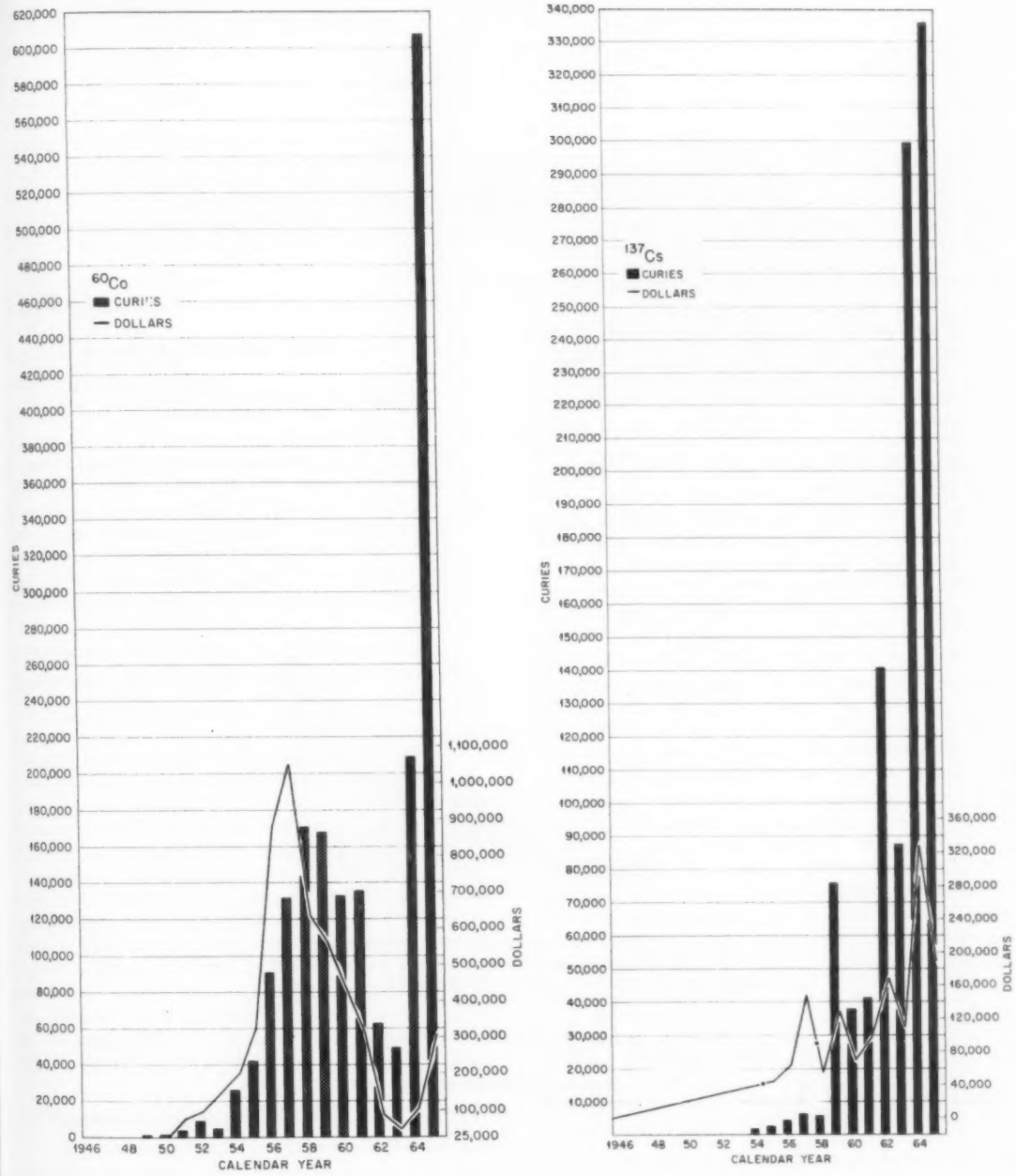


Fig. 2 Radioisotopes for large-scale irradiations.

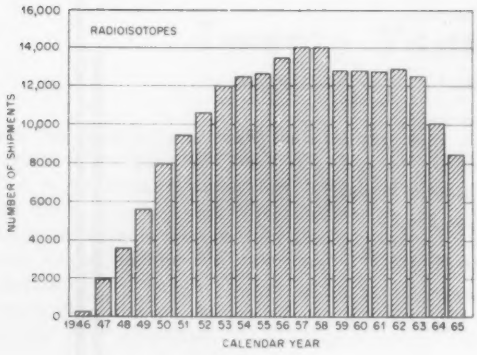


Fig. 3 Shipments of radioisotopes from ORNL, 1946–1965.

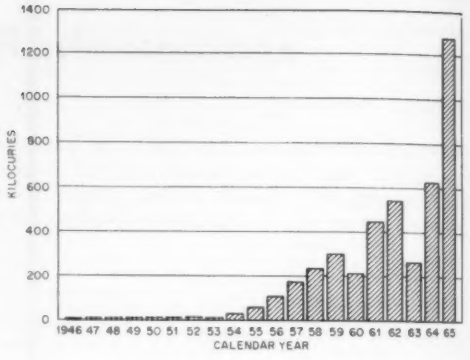
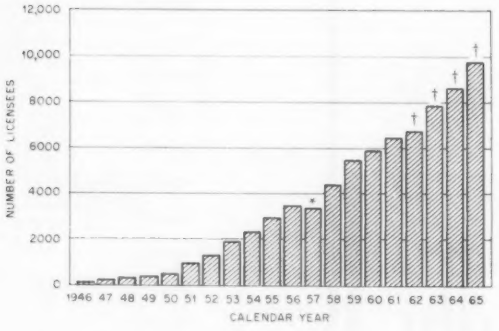


Fig. 4 Curies of radioisotopes shipped from ORNL, 1946–1965.



*Prior to 1957, the summary total of licensees was obtained by adding new licensees to the total from the previous year. In 1957 and subsequently, the totals include only valid licensees for the current year. The drop in 1957 reflects the culling process.

†Includes licensees in "agreement states."

Fig. 5 Radioisotope licensees, 1946–1965.

Miscellaneous

Conferences

Symposium on Mössbauer Effect in Chemistry

A 1-day symposium on the Mössbauer effect and its application in chemistry was sponsored by Nuclear Science and Engineering Corporation in New York, Sept. 12, 1966. Five papers were presented on Mössbauer spectroscopy of specific elements, and five papers covered structural studies, studies of magnetic properties, coordination chemistry in biological compounds, ordering phenomena and oxidation, and an introduction to Mössbauer spectroscopy.

Symposium on Tracers in Industry and Geophysics Held in Prague, Czechoslovakia

A tentative program has been received for the Symposium on Radioisotope Tracers in Industry and Geophysics to be held in Prague, Nov. 21–25, 1966. Session I will consist of 2 papers on industrial applications of radioisotope tracers. Session II will contain 6 papers on friction and wear studies. During Session III, 6 papers will be presented on geophysical applications. Seven papers will be given during Session IV on dispersion of effluent and miscellaneous applications. For the last $2\frac{1}{2}$ days of the conference, papers will be presented on plant kinetics, where "plant" refers solely to "factory." About 25 papers are listed under this category. Representatives of 18 countries are scheduled to present papers. The proceedings will be published by the International Atomic Energy Agency (IAEA).

XI Nuclear Congress of Rome

At the eleventh Nuclear Congress of Rome, held June 23–25, papers were presented on both production and industrial use. H. Sheard of Amersham, U. K., pointed out that power reactors will be used for producing only a limited number of radioisotopes, e.g., ^{60}Co, ^{237}Np, ^{238}Pu, and emphasized that power-reactor neutrons are not really "free." J. Ransohoff of Neutron Products, Washington, D. C., discussed the elementary economics of radioisotope production in power reactors; his main point was that financial loss as well as profit is a possible result of engaging in this field. He also discussed the joint program of his company and Consumers Power Company to produce several megacuries of ^{60}Co in the utility's reactor at Big Rock Point. (See other article in this section.) C. Fisher of the Commissariat à l'Energie Atomique (CEA), Saclay, France, and U. Rose of Centro Ricerche Nucleari (SORIN), Sallugia, Italy, spoke on their radioisotope production programs, which are associated with that of Belgium, in a common radioisotope production and marketing combine. The total CEA–CEN–SORIN establishment has 200 production people and 9 reactors; they make 600 products, and the 1965 income was the equivalent of nearly $2 million. Their output in 1965 for some important isotopes was: ^{131}I, 459 curies; ^{198}Au colloid, 152 curies; ^{32}P, 114 curies; ^{192}Ir, 13,000 curies; ^{60}Co, 6000 curies. T. Slosek of General Electric Company, Pleasanton, Calif., speaking on commercial radioisotope production in the United States, said that General Electric plans to raise the power of the Vallecitos Experimental Superheat Reactor from 33 to 50 Mw, giving them fluxes up to 5×10^{14} n/(cm^2) (sec). General Electric is also studying the possibilities of building a 100-Mw reactor optimized for radioisotope production. T. S. Weismann of Isochem, Inc., Richland, Wash., described their Richland fission-product plant project and indicated that they would be in operation in 1968.

AEC Activities

DID Director E. E. Fowler Announces Top Assistants

E. E. Fowler, head of AEC's Division of Isotopes Development (DID), has announced the appointments of G. Y. Jordy and W. E. Mott as his top aides. Jordy, who has been with the AEC since 1957, will be assistant director for planning and evaluation. Mott will act as assistant director for technical programs.

Isochem Applies for Permit To Build Plant at Hanford

Isochem, Inc., a subsidiary of United States Rubber Company and Martin Marietta Corp., has applied to the AEC for a permit to construct a Fission Products Conversion and Encapsulation Plant at the AEC's Hanford Works near Richland, Wash., to begin commercial operation in late 1968. The application estimates cost of construction at $8 million, although this estimate has "grown" considerably since the application was filed. The plant will recover ^{90}Sr, ^{137}Cs, ^{147}Pm, and ^{144}Ce from waste products of the chemical processing facilities at Hanford, then process them for sale. This will be the first privately owned large-scale commercial facility for treatment of these fission products.

NUMEC Winner of Heart Pacemaker Contract

The AEC has announced that Nuclear Materials and Equipment Corp. has been awarded a contract to develop an isotope-powered cardiac pacer. The AEC predicts costs of from one-half to three-quarters of a million dollars. The contract covers design, development, and engineering of the pacers; overall guidance will be provided by the National Heart Institute of the National Institutes of Health and by Beth Israel Hospital, Newark. The isotope selected is ^{238}Pu; about 0.6 g of the isotope will be used.

As with conventional battery-powered cardiac pacers, the nuclear device will be implanted in the chest cavity by surgery. It is expected to have a minimum operating life of 10 years.

[A full description of AEC plans for the cardiac pacers was given in *Isotopes and Radiation Technology*, 3(3): 293-294 (Spring 1966).]

ORNL Has Two Big Events: One New and One Old

In early September two significant events occurred at Oak Ridge National Laboratory (ORNL): First, on September 9 the High Flux Isotope Reactor (HFIR) reached full power for the first time. At this 100-Mw level, the thermal flux in the trap region is 5×10^{15} n/(cm^2)(sec) and exceeds that in any other research reactor. The HFIR is to be used for production of the first gram quantities of californium, but will also be used for basic research as well as for radioisotope production studies.

Second, on September 13, in the presence of several hundred persons, the world-famous Oak Ridge Graphite Reactor, the oldest of production reactors, was dedicated as a "National Historic Landmark" by the National Park Service (NPS) of the U. S. Department of the Interior, in recognition of its 20 years of yeoman service (1943–1963)—first, to produce small but weighable amounts of ^{239}Pu to test Dr. Seaborg's separation methods and to acquaint the future operators of the large Hanford reactors with nuclear energy and nuclear radiation; second, to develop neutron-diffraction techniques and demonstrate nuclear alignment and the radioactive decay of the neutron; and third, to produce radioisotopes for science and medicine. Presentation of the plaque and certificate was made by Mr. Elbert Cox, Regional Director of the NPS, to Dr. Glenn T. Seaborg, accepting in behalf of the AEC. He, in turn, presented them to Dr. A. M. Weinberg, Director of ORNL. Those making remarks at the ceremony were Cox, Seaborg, Weinberg, and Dr. Eugene P. Wigner, former Research Director at ORNL and one of the men who provided the guidance and inspiration to those who carried out the detailed design of the X-10 Reactor. Dr. Samuel M. Nabrit, new AEC Commissioner, was among the celebrities attending the dedication.

AEC Announces Modifications to Container Labeling Requirements

In the future more information will be required on containers holding radioactive materials licensed by the AEC. Above specified quantities, the radioactive material will need a label that states the level of radiation, kind of material, amount of material, date for which the radioactivity is estimated, and enrichment or percentages of the material. This applies to shipping containers as well as storage containers.

Exceptions are containers that are attended by an individual who takes the precautions necessary to prevent overexposure of persons around the container; containers in transport that comply with regulations of the Interstate Commerce Commission, Federal Aviation Agency, or Coast Guard; containers accessible only to individuals qualified to handle, use, or work near them; and manufacturing or process equipment already covered under more specific controls.

The amendments to AEC regulations, Title 10, *Code of Federal Regulations*, Part 20, "Standards for Protection Against Radiation," will become effective 90 days after publication in the *Federal Register* on Aug. 5, 1966.

Highest Specific Activity ^{60}Co Produced at Savannah River

The Savannah River Plant in Aiken, S. C., has made the most concentrated ^{60}Co source in the world (see Fig. 1). The source consists of two small nickel-plated wafers of ^{59}Co containing ^{60}Co, weighs 1.4 g, and has an activity of 980 curies—or 700 curies/g. Eight months' irradiation at high flux in one of the high-flux reactors at the Du Pont-operated plant was necessary to achieve this high specific activity.

The highest concentration previously achieved was 405 curies/g in a ^{60}Co source produced at the National Reactor Testing Station.

Fig. 1 The most concentrated ^{60}Co source in the world. The 700 curie/g source is stored under 10 ft of water in a cooling basin after being removed from the reactor.

AEC Approves Agreement with Alabama

Alabama became the 13th state to reach an agreement with the AEC on the control of radioactive materials in the state. The transfer of regulatory authority will include licensing, rule making, and enforcement in uses of radioisotopes, source materials uranium and thorium, and small quantities of fissionable materials. The Division of Radiological Health of the Alabama Board of Health will be the regulatory agency. There are about 205 AEC licenses in Alabama.

Other states that have reached similar agreements with AEC are: Arkansas, California, Florida, Kansas, Kentucky, Mississippi, New Hampshire, New York, North Carolina, Oregon, Tennessee, and Texas. Agreements with Louisiana and Nebraska are pending.

Recent Large ^{60}Co Shipments from ORNL

More than 2.5 Mc of special shipments of ^{60}Co have been handled by ORNL this year, over 475,000 curies of which had been sent to the Canadian AECL Chalk River Nuclear Laboratories by August 26. Another large shipment is the 1.4 Mc being shipped to Babcock & Wilcox Co., Lynchburg, Va., for encapsulation and eventual use in the U. S. Army Natick Laboratories food irradiator. Also being handled by ORNL is a shipment of 790,000 curies sent from Brookhaven National Laboratory to Lockheed-Georgia, Dawsonville, Ga., for encapsulation.

Availability of Isotopes

ORNL Stable-Isotope Inventory Augmented

Recent additions to the ORNL enriched stable-isotope inventory include the following:

Isotope	Purity, %	Amount available, g
^{54}Fe	97.1	60.0
^{70}Ge	98.8	3.8
^{72}Ge	98.7	5.3
^{91}Zr	91.8	25.0
^{96}Zr	85.2	1.8
^{98}Mo	98.8	65.0
^{113}Cd	95.0	23.0
^{125}Te	95.5	6.6
^{126}Te	98.6	15.0
^{138}Ce	14.3	4.7
^{140}Ce	99.7	400.0
^{196}Hg	49.1	0.3
^{204}Hg	80.9	0.4

General

New Half-life Value for ^{147}Pm

Since publishing the table of revised half-life values in *Isotopes and Radiation Technology*, 3(3): 294 (Spring 1966), the Isotopes Development Center (IDC) has received notice from ORNL's Nuclear Data Group that the half-life of ^{147}Pm is 2.62 years instead of 2.65 years. This new value contains some late-1965 data and is now being used by the IDC.

Army Asks Clearance for Irradiated Ham

The U. S. Army, in a petition filed July 11, asked for U. S. Food and Drug Administration (FDA) approval of consumption of irradiated canned ham. The use of a 3.5- to 5.6-Mrad dose from ^{60}Co or ^{137}Cs will permit storage of the ham for a year or more without refrigeration. Since unirradiated ham has to be refrigerated until eaten, the Army believes a favorable decision will lead to ultimate commercialization of the process in the United States.

Petitions for clearance of radiation-processed strawberries, oranges, and six kinds of fish are already before the FDA, which has so far cleared irradiated bacon, potatoes, and wheat. FDA action usually takes a year or more.

Union Carbide Becomes Fifth Radiopharmaceutical Firm

Through Neisler Laboratories, a subsidiary purchased last year, Union Carbide Corp. has become a radiopharmaceutical firm. Under the name Neimotec, a ^{99m}Tc generator is being offered in strengths of 100, 150, and 200 mc. Since this "cow" is not refined, sterilization is necessary after elution. Union Carbide hopes to have two radionuclide solutions for sale by Neisler in November and is stressing the fact that it is the only integrated producer–supplier in the business.

New Promotional Brochures Available from Industry

A full-color brochure on Novawood is now available from The American Novawood Corporation. The four-

page pamphlet explains what Novawood is (an irradiated wood–plastic combination) and lists current products of the firm. For a copy, write: The American Novawood Corporation, 2432 Lakeside Drive, Lynchburg, Va. 24501.

High Voltage Engineering Corp. (HVEC) is now publishing an occasional newsletter. Volume 1, No. 3, was published July 22, 1966. The current issue lists current projects of HVEC and touts successful applications of the firm's accelerators. Other items of interest to accelerator users are included. For copies write: High Voltage Engineering Corp., Burlington, Mass. 01803.

IAEA Panel of Experts from Six Nations Emphasizes Small Atomic Batteries

Representatives from Austria, Federal Republic of Germany, France, Sweden, United Kingdom, and United States recently met in Vienna to consider terrestrial applications of isotope-powered sources of electricity. This panel concluded that the devices are now practical for uses for shipping, meteorology, communications, medicine, and other purposes.

A large amount of knowledge has been acquired on isotope-powered thermoelectric devices as a result of their application to the space program, especially in the United States. Quantities of cheap isotopes are becoming available, and there are potential applications of which the appropriate authorities may not be aware. Their primary application to date has been as a power source in remote areas, e.g., weather stations and lighthouses. At the other end of the scale is their use in medicine as "pacemakers" for various organs in the body, such as the heart. Other possible uses include warning devices for seismic, flood, and forest-fire purposes.

The isotopes usually used are ^{90}Sr and ^{238}Pu. The panel felt strongly that regulations for handling and transport and for standardization of sources should be handled by the IAEA.

Deering Milliken Announces New Fabric Developed with Electron Radiation

At its 13th Annual Milliken Breakfast Show, Deering Milliken & Co., Inc., announced the successful application of electrons to textiles in a new exclusive process that results in a fabric which has superior soil-releasing properties. Washed under normal home-laundry conditions with ordinary detergents, VISA will release even stains left by butter or salad oil. The fabric, made of 65% Dacron polyester and 35% cotton, is postcured and has a high-quality durable press. A similar fabric, MILSTAR, with the same quality durable press but without the soil-releasing quality of VISA, has also been developed with the use of electron radiation.

Mid and heavy weights in VISA and light, mid, and heavy weights in MILSTAR will be manufactured and marketed for men's, women's, and children's clothing. Deering Milliken has equipped Magnolia Finishing Plant (Blacksburg, S. C.) with several 500-kv electron accelerators, and the fabric is now being produced in quantity.

Sulzer Brothers Ltd., Winterthur, Switzerland–Four Irradiator Contracts at Once

Sulzer Brothers, Ltd., of Winterthur, Switzerland, currently has four big cobalt installations under construction or on order. For B. Braun of Melsungen, Germany, Sulzer is building a fully automatic gamma irradiation plant for the sterilization of catgut and other medical and pharmaceutical supplies. For Farbwerke Hoechst, Frankfurt-am-Main, Germany, Sulzer is building an irradiation plant for chemical research work. The Technical University of Munich has ordered a gamma irradiation plant as part of a radiochemical institute being built as part of the research center at Garching, near Munich. An unnamed client has ordered a plant similar to the Farbwerke Hoechst plant.

The Braun medical irradiator is designed for a maximum capacity of 600,000 curies of ^{60}Co, but the initial load will be only 30,000 curies. The packed sutures will be delivered by a conveyor, through a labyrinth, to the individual source rods arranged on a stand. The operation is fully automatic.

The chemical research irradiator for Farbwerke Hoechst contains two identical units that will operate independently; they are designed for a maximum of 10,000 curies each; the total initial load will be 11,000 curies. Each source is stored in the ceiling of its cell, and each cell has a separate control desk. Special attention was given to easy operation of the devices used to guide as well as store the sources.

The unit ordered by the Technical University of Munich is an open-room irradiator with a maximum capacity of 100,000 curies of ^{60}Co. The initial loading will be 10,000 curies. The source is made up of eight rods in a circle, with the diameter of the circle varied by an iris arrangement.

The 650,000-curie irradiation plant for the anonymous purchaser is similar to the Farbwerke Hoechst plant but will use ^{137}Cs.

India Obtains Two ^{60}Co Irradiators from Canada

Two irradiators for experiments in food preservation and grain disinfestation have been ordered by the Indian Department of Atomic Energy. The order has been placed with Atomic Energy of Canada Ltd. One source will be at least 100,000 curies; the other will be a portable irradiator of approximately 28,000 curies. Both irradiators were discussed at the International Symposium on Food Irradiation at Karlsruhe, Federal Republic of Germany, in June 1966.

The original load of ^{60}Co will come from Canada, but further loads will probably be provided by India from her own reactors.

Irradiation of Organic Coating—Dynacote Vs. Ford Electrocure

Two different processes, one patented, have recently been publicized on the radiation treatment of thin organic coatings. Radiation Dynamics, Inc., Westbury, L. I., is marketing a 300-kev electron accelerator. The company says the new accelerator is designed to cure paint, lacquer, and dye coatings. Styrenated polyesters, polyvinyl chloride plastisols, acrylics, epoxy acrylics, styrenated polyester plastisols, and a variety of coatings can be treated. No solvent is needed.

Ford Motor Company has been granted U. S. Patent 3,247,012, covering coating articles with radiation-polymerizable coatings. Ford has also applied for a patent on modifications to a commercially available accelerator. The Ford process is outlined in a small brochure available from the Manager of the Electrocure Department, Ford Motor Company, American Road, Dearborn, Mich. Ford's first licensee is Boise Cascade Corp., which will use the process to coat wood and wood products.

High-Power Industrial Radiation Facility To Use Electron Accelerators

Electra-Process Industries plans to build what it is billing as "the world's most powerful industrial radiation facility" near Spokane, Wash. The $700,000 plant will be built primarily to sterilize meat, but it can be used to treat other foods and also medical supplies. Radiation services will be provided. The plant will use high power levels of 60 to 90 kw, with high-energy electrons (7 to 15 Mev) for deep penetration and high rate of throughput. Mid-1967 is the target for production.

Neutron Products Gives Details of Big Rock Point Cobalt Production

The large-scale production of ^{60}Co was initiated on May 9, 1966, in the Big Rock Point Reactor at Charlevoix, Mich., under a joint program between the owner, Consumers Power Company, and Neutron Products, Inc. J. A. Ransohoff, president of Neutron Products, predicts production of 450,000 curies per year; by late 1967, 2 Mc will be produced. The cobalt will be encapsulated in 304L stainless-steel tubes, 12 in. long and 0.271 in. in diameter. Standard marketing classes are shown in Table 1.

Table 1 STANDARD CLASSES OF NEUTRON PRODUCTS ^{60}Co

Class	Nominal specific activity, curies/g	Curies per rod	Double encapsulated source plaque, curies/sq ft	Estimated availability date of kilocurie amounts
D	20	700	28,000	May 1967*
C	30	1050	42,000	May 1967
B	40	1400	56,000	November 1967†
A	50	1750	70,000	November 1967
Medical‡	60–80	>1800	-	November 1967

*Limited quantities may be available in November 1966.
†Limited quantities may be available in May 1967.
‡Deliverable as once-encapsulated wire convertible into pellet form.

Isotope Fluorescence Analyzers in Field and Laboratory Models

The isotope fluorescence analyzers reported in *Isotopes and Radiation Technology*, 3(2): 95 (Winter 1965–1966) and now available in the United States. Hilger and Watts Ltd. of London has authorized Engis Equipment Co. of Morton Grove, Ill., to sell both models.

The portable isotope fluorescence analyzer (PIF) is intended for analysis of ores in situ; it can analyze metallic ore to within 0.05% in only 30 sec. A small button-shaped radioisotope source about 1 cm in diameter emits radiation to a target sample (for example, a rock face in a mine), and the back-scattered radiation passes through a pair of X-ray filters chosen to allow only the specific X-ray emission of the wanted element to pass through. Two 6-volt dry batteries supply power to the electronic control unit, which is carried by a shoulder strap. The whole device weighs less than 20 lb. The radioisotope used currently for tin is 500 mc of ^{147}Pm.

The laboratory model, the LIF analyzer, can be used in rolling mills, factories, and elsewhere whenever there is a need to ensure that material specifications are exact. It is similar in operation to the PIF analyzer, but it can operate from a main power supply as well as its own batteries. The radioisotope is selected to suit the use; so far, ^{147}Pm, ^{3}H, and ^{109}Cd have been used.

Westinghouse Receives Navy Contract To Develop Nuclear Battery

Westinghouse Electric Corp., Pittsburgh, has been awarded a contract by the U. S. Navy to develop a 5-watt thermoelectric generator based on ^{90}Sr. The device must operate underwater unattended for 5 years at depths of 20,000 ft and at pressures of 10,000 psi.

The Westinghouse nuclear battery is compact, 24 in. in diameter and 29 in. long. It will use a new tubular design of concentric cylinders; the heat source can be in the generator core or can be external. The Navy has requested an external mounting of ^{90}Sr.

Westinghouse plans to deliver the device by the end of the year to the Naval Civil Engineering Laboratory in Port Hueneme, Calif., for evaluation.

First Nuclear Generator in Space Celebrates Fifth Anniversary

On June 29, 1966, the first nuclear-powered generator began its sixth year in space. It is still

providing power to the Navy's experimental navigational satellite 4-A, the oldest operating U. S. satellite. The historic generator has now traveled over 25,000 times around the earth, more than 724 million miles. The Martin Co., Baltimore, was the prime AEC contractor for the SNAP device.

The aim of the SNAP program is the development of compact, lightweight, reliable nuclear electric devices that will operate unattended in all environments. More advanced versions are being developed.

Auto Manufacturers Evaluate Radioluminescent Devices

A recent publicity release from Minnesota Mining & Manufacturing Co. states that self-lighting radioisotope devices are being evaluated by the leading automobile manufacturers for use in windshield-wiper knobs, gearshift indicators, ignition key locks, and door keyholes. The radioisotopes used are the beta emitters ^{3}H and ^{147}Pm. The devices are reliable, permanent, and less expensive than currently used electric-bulb systems. While manufacturing the devices requires a general license from the AEC, using the devices involves no regulations.

Unique Opera House Uses Tracers To Determine Possible Faults

Ove Arup & Partners, Consulting Engineers to the Sydney (Australia) Opera House Project, in December 1965 employed Unisearch Ltd., the research and development company of the University of New South Wales, to discover flaws in the poured concrete roof of the unfinished structure with the use of radioactive tracers.

Labeled sodium carbonate isotope (^{24}Na) with a half-life of 15 hr was pumped into large sections of the building still under construction. Mixed in a shielded tank, the radio-labeled liquid was pumped up (rather than down, because of the access problems—see Fig. 2) between the strands of the steel hawsers which run through the middle of the dense concrete shells of the Opera House roof. Scientists with portable scintillation counters then climbed into the scaffolding to look for locations of increased radioactivity. These locations would occur at openings or voids in the concrete.

The stems forming the underpart of the concrete roof shells were investigated also; transmission of gamma rays from ^{137}Cs was used in this investigation.

Martin Program of Commercial SNAP Devices Gains Momentum

In the spring of 1966, the Martin Co., Baltimore, a division of the Martin Marietta Corp., announced that it was releasing for sale 25-watt nuclear generators using ^{90}Sr. Eight models are being offered, four each for terrestrial and for underwater use. A generator with no warranty will cost $53,587; a generator with a 2-year warranty on workmanship and materials will cost $55,400. A 2-year guarantee of the power will raise the price to $57,680; a 5-year guarantee of the rated power will cost $63,230.

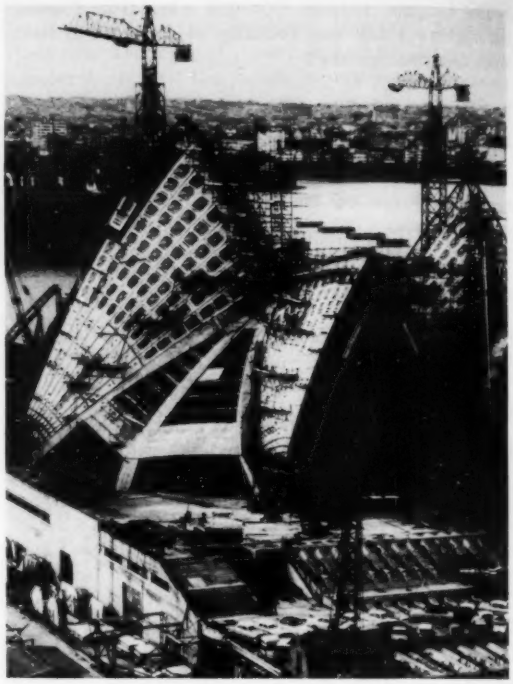


Fig. 2 The Sydney Opera House under construction, showing the massive concrete structures.

Company officials have said they expect sales of $4 to $5 million in 1966. The nuclear batteries will be useful in remote areas for radio communication, for navigational aids, and for automatic weather stations.

The U. S. Navy was the first customer. The Naval Electronics Laboratory bought a 25-watt model warranted for 2-years' output and installed it on a small island in the Bering Strait to power an oceanographic measuring station. No other sales have been reported.

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